LA-UR-93-4373

CHARACTERIZATION OF THE RADIOACTIVITY IN SURFACE-WATERS AND SEDIMENTS COLLECTED AT THE ROCKY FLATS FACILITY

D. W. Efurd, D. J. Rokop, and R. E. Perrin

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SW-A-004568

INFORMAL MEMORANDUM

TO: Bob Stevens EG&G SWD, T893A
CC: G. Hill, DOE ES&H EPD, T117
S. Pettis, EG&G SWD, T893A
FROM: R.D. Lindberg, EPD SMS
DATE: February 4, 1994

SUBJECT: **Review of Characterization of the Radioactivity in Surface Waters and Sediments Collected at the Rocky Flats Facility**

This is an informal technical review of the LANL report by Efurd, Rokop and Perrin, LA-UR-93-4373 regarding nuclide characterization work for EG&G's Surface Water Division. This informal review was requested by S. Pettis.

General Comments.

1. Overall the report is well written and clearly conveys the work that was done.

2. I would like to see this LANL team do a similar study on our groundwater for the Geosciences group (S. Singer, heads the groundwater program). I will talk with Singer about scoping/funding a small project. In general Rocky Flats groundwater is more contaminated with metals and nuclides than anything LANL analyzed in surface water. It will be interesting to see what the U-236 activities are some of our wells. I would like to talk with LANL about analyzing for other nuclides (like those in Table XIV) and Np and Cm isotopes used at RFP. We also analyze for fallout Cs-137, but not for Cs-134 which has a state groundwater standard!

3. The assumption throughout this report is that Rocky Flats Plant (RFP) only has a depleted U contribution superimposed on naturally occurring background uranium. In my opinion, the reality is that RFP used U of enriched, depleted and natural isotopic compositions, and these contributions are superimposed on the naturally occurring background uranium. The Solar Evaporation Ponds (SEP) have undoubtedly received low level process wastes with U of all three isotopic compositions at various times. (Your enriched U sample of OU4 tank water confirms this on page 46). Leakage of these ponds (first observed in 1954) has created a large groundwater plume immediately upgradient of Pond A1. There is some indication of "residual groundwater contamination" below Pond A4. The possible mixing between pond waters and alluvial groundwater in the A and B series ponds has never been characterized. There is evidence that groundwater contamination plumes have short travel times (on the order of 5 years) through the valley fill alluvium to the eastern plant boundary at Indiana. If all three U compositions were released to the Walnut Creek drainage then the binary mixing model discussed on page 26 is invalid, and therefor the results shown as Table VI and Table XIII are suspect. At DOE direction, EG&G Geosciences Division is presently planning a detailed data collection and groundwater fate and transport modeling effort to quantify plume movements and groundwater-surface water interactions.

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Specific Comments

Page. 2. I disagree with the statement that 20% of the U in Pond B-5 originated as depleted U, per general comment 3 above.

Page. 17. An English major might object to this entire page being one long paragraph, but I wish to ask about a statement three sentences up from the bottom. Please provide some examples of the insoluble Th, Pu, Am, Po and Bi products that concentrate in the sediments. My work suggests that Am is highly mobile in RFP groundwaters.

Page 26. I could not duplicate the percentages shown on Table VI by using your stated assumption that depleted U contains 0.5% U-235. I back-calculated a few of LANL's values and come up with an atom-ratio of 220 for U-238/U-235. This translates to 0.45% U-235. The difference is not a big deal, but the text is incorrect. More importantly, LANL should have used the actual average composition of RFP "depleted" U rather than the national average! The 1980 RFP EIS (p.2-172) gives some numbers. If I did the calculation correctly these numbers translate to 0.23% U-235 in typical RFP depleted uranium. Therefore your Table VI percentages are probably off, even if you are correct in using the "binary" model.

Page 32-33. The Pu-240/Pu-239 ratio work is very nice, and the results no surprise.

Page 33 next to last sentence. The Pu content of your surface water samples may be higher than the Am because of surface runoff. This is not true in RFP groundwater where it is common to find the highly mobile Am 5 times to 10 times higher in activity than the Pu (which gets left behind in the soil environment).

Page 33, 2nd sentence, last para. Clarify and provide a reference for your statement that the alpha activity ratio of "materials processed at RFP" ranges from 0.2 to 0.4.

Page 38 and 39 table. Here and elsewhere in the text LANL refers to the "Walnut and Indian" sampling location. I'm sure EG&G has pointed out that this should be "Walnut and Indiana".

Page 42, first sentence of last paragraph. Typo, change "collects" to "collected".

Page 43, last paragraph. Please specify what the "expected" fallout concentrations are and give a reference.

Page 44, top. I would like to see the predicted activity ratios assuming 100% natural U and Th. Also, the last sentence of the first paragraph covers all possibilities, adds nothing, and can be deleted.

Page 46. I know you use atom percent because it falls out of the mass spec work, but for the rest of us please make some conversions to pCi/l.

Page 47, next to last paragraph starting with "All". Sorry this statement covers all possibilities and adds nothing to our understanding. I would drop it.

Page 49, suggestion 6. You should believe your instruments. We <u>do</u> have enriched U at RFP!

P.S. Let's talk! I think we might have some work for LANL characterizing nuclides in RFP groundwater.

CHARACTERIZATION OF THE RADIOACTIVITY IN SURFACE-WATERS AND SEDIMENTS COLLECTED AT THE ROCKY FLATS FACILITY

FINAL REPORT

WP:61205;WATER CHARACTERIZATION [LATO-EG&G-91-022 (FY93)]

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EXECUTIVE SUMMARY

This study was initiated to characterize the radioactivity in surface-waters and sediments collected at the Rocky Flats Plant (RFP). The study quantified the amount of radioactivity present and determined whether the radioactivity was naturally occurring or anthropogenic. Another objective of the study was to analyze samples collected from locations that may contain radioactive sources that could increase the radioactive surface-waters inventories at RFP. The data collected during this study serve as a base-line by which the impact of future remediation efforts can be evaluated. Eighty-three water samples and 24 sediment samples were collected in support of the project. Over 800 separate analyses were performed. Waters in the terminal ponds A-4, B-5, C-2 and the effluent from the Sewage Treatment Plant (STP) were sampled monthly. The waters in Ponds A-1, A-2, A-3, B-1, B-2, B-3, B-4 and C-1 were sampled quarterly. Special soil, sediment and water samples collected at locations that could affect the surface-waters radioactivity inventories at RFP were also analyzed.

The largest source of anthropogenic radioactivity detected during this study was the sediments currently residing in the ponds. One gram of sediment from a holding pond contains approximately 50 times more plutonium than 1 liter of water from the pond. Two other specific locations have been identified that may contribute to the radioactive surface-waters inventories at RFP. Plutonium and depleted uranium appear to be moving down the South Interceptor Ditch and through the A-1 Bypass.

The largest source of radioactivity in the terminal ponds was naturally occurring uranium and its decay product radium. There is 70-450 times more alpha activity resulting from the decay of naturally occurring radium than alpha activity resulting from the plutonium in the terminal ponds. The largest source of anthropogenic radioactivity in the terminal ponds was depleted uranium. Approximately half of the uranium present in Ponds A-4 and C-2 originated as depleted uranium. Approximately 20% of the uranium in the waters collected from Pond B-5 originated as depleted uranium.

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INTRODUCTION

The Rocky Flats Plant (RFP) is a Department of Energy (DOE) facility where plutonium and uranium components were manufactured for nuclear weapons. During plant operations radioactivity was inadvertently released into the environment. Plutonium has been detected in air, soil and water samples at RFP. The largest single contributor to plutonium in the environment is resuspension of contaminants originating at the OU2/903 Pad.[1] Between January and June 1968 oil drums containing cutting oil contaminated with plutonium were removed from the OU2/903 Pad. Some of the drums leaked and high winds spread the plutonium. Seed, *et al.* estimated that 6.1 Ci of plutonium were lost to the soil.[2] Uranium releases have also occurred. The amount of uranium released by RFP has not been studied as thoroughly as the plutonium releases. Actinides may be introduced into surface-waters at RFP by inadvertent release of contaminated fluids from the plant site, wind deposition of contaminated soil and erosion of topsoil by rain and snow melt. A portion of the surface-water collected at RFP is discharged off site. This provides a potential pathway to transport plutonium and uranium into reservoirs that serve as public water supplies. This study was initiated to characterize the radioactivity present in surface-waters at RFP. The naturally occurring and anthropogenic radioactivities were measured in water, sediment and soil samples.

SURFACE-WATER HYDROLOGY

Figures 1 and 2 illustrate the creeks, diversion ditches, ponds and reservoirs in the RFP vicinity. Their functions are described below. Three drainage basins and natural ephemeral streams transverse RFP. The Woman Creek drainage basin traverses and drains the southern portion of the site. The Rock Creek drainage basin drains the northwestern portion of the plant complex. The Walnut Creek drainage basin traverses the western, northern, and northeastern portions of the RFP site. Three ephemeral streams are tributary to Walnut Creek. The streams are Dry Creek, North Walnut Creek and South Walnut Creek.

Figure 2 gives a schematic representation of the surface-water systems at RFP. Dams, detention ponds, diversion structures, and ditches have been constructed at RFP to control the release of plant discharges and

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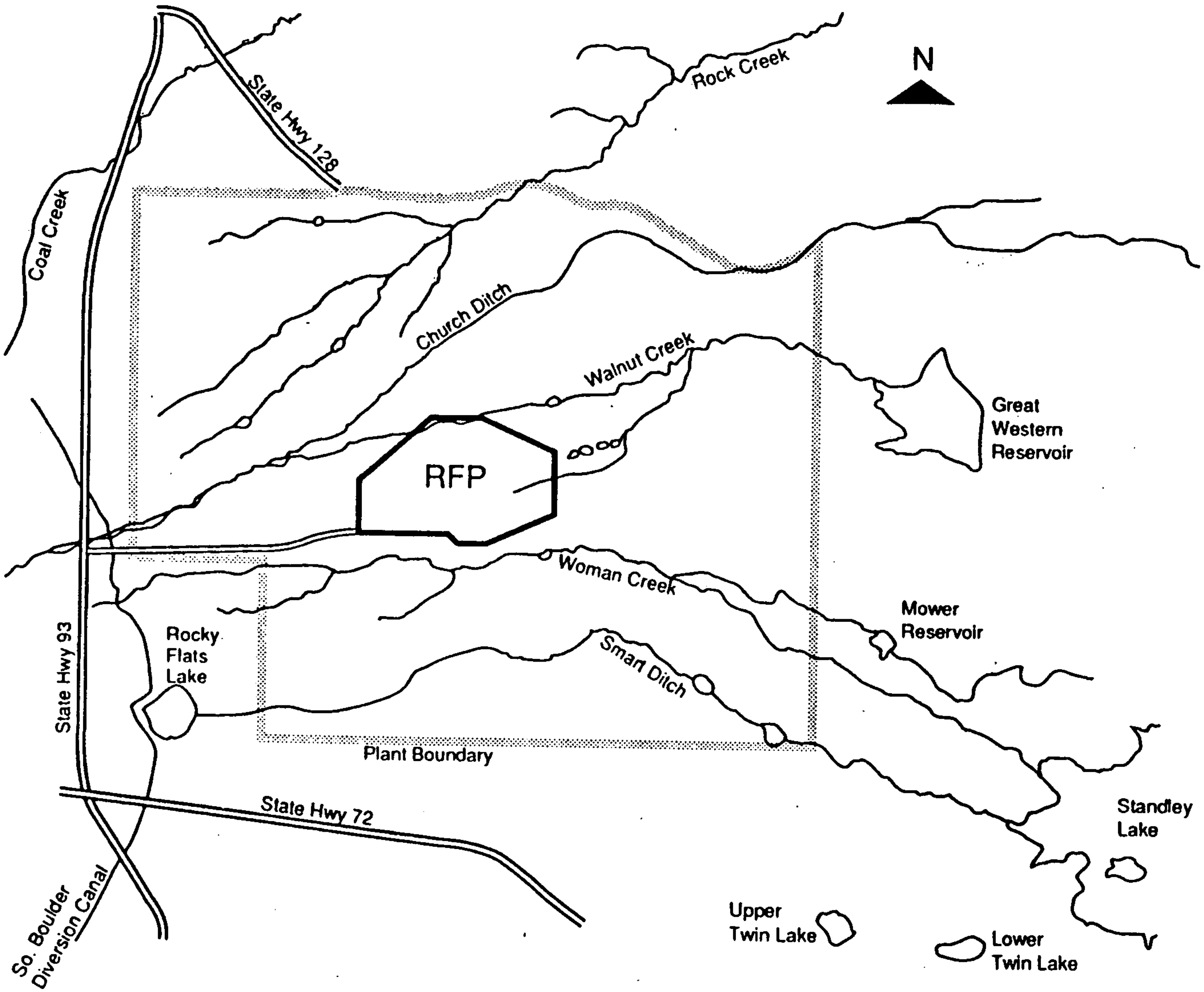
Rocky Flats Plant and Surrounding Area Drainage



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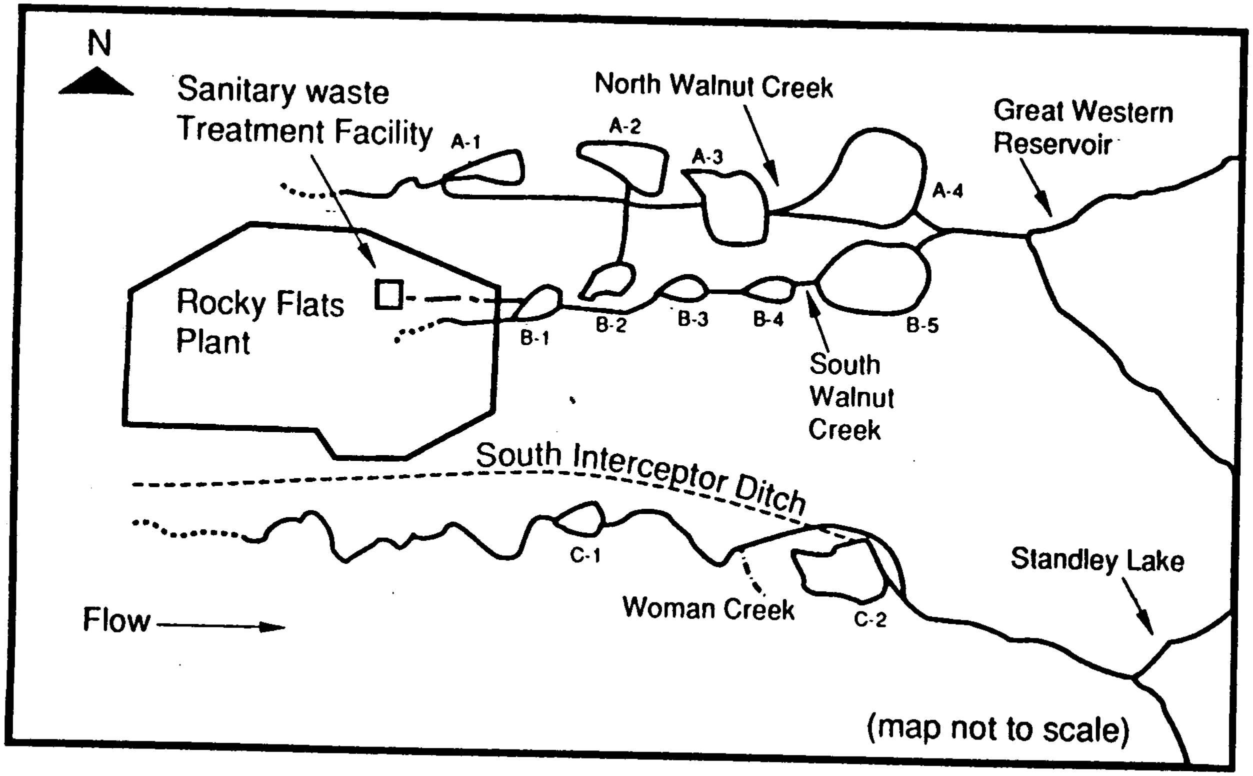
Detailed Schematic of Rocky Flats Plant Pond System



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surface (storm water) runoff. The ponds located downstream of the plant complex on North Walnut Creek are designated A-1 through A-4. Ponds on South Walnut Creek are designated B-1 through B-5. The ponds in the Woman Creek drainage basin are designated C-1 and C-2.

Ponds A-1 and A-2 were constructed during the early days of plant operation. These ponds are currently used to collect seep and culvert flows and precipitation runoff from the northern area of the plant site. Ponds A-1 and A-2 are now being operated as non-discharge ponds. Volumes of water are controlled by overpond spray evaporation. Pond A-3 collects surface-water diverted around Ponds A-1 and A-2 and a substantial portion of the North Walnut Creek and northern plant site runoff. Pond A-3 is operated in a detain, sample, analyze and release mode. Water released from Pond A-3 is transported to Pond A-4. Pond A-4 was constructed in the 1980s.

Ponds B-1 and B-2 were constructed during the early days of plant operation. Ponds B-1 and B-2 are currently used to collect suspect flows or upsets from the sewage treatment plant (STP) operated at RFP. These ponds are currently operated as non-discharge retention ponds. Water from Ponds B-1 and B-2 is transferred to A-2 after characterization. Pond B-3 receives treated effluent from the STP. Approximately 10-15% of the water treated by STP originates from miscellaneous industrial sources. Water from Pond B-3 is released into Ponds B-4 and B-5. Pond B-4 is operated as a flow-through pond. Pond B-5, constructed in the 1980s, is a terminal pond.

Pond C-1 is located on Woman Creek and receives natural flows. An interceptor ditch (South Interceptor Ditch) is located between and parallel to Woman Creek and the southern boundary of the plant complex. The South Interceptor Ditch drains into Pond C-2 which was constructed in the 1980s. Any offsite discharge from RFP is from the terminal ponds (Ponds A-4, B-5 or C-2).

One retention pond (the Landfill Pond) is located in an unnamed basin immediately down gradient of the present Landfill. The Landfill Pond was constructed in 1974. It is operated in a zero discharge mode through spray evaporation.

In addition to natural flows and the South Interceptor Ditch, there are seven ditches or diversion canals in the general vicinity of RFP. The

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Upper Church, McKay, Kinner, and Reservoir County Ditches cross the site. The Upper Church Ditch delivers water to Upper Church Lake and Great Western Reservoir. McKay Ditch supplies water to Great Western Reservoir. The Great Western Reservoir is a water supply for the City of Broomfield. Until recently water from Walnut Creek also drained into Great Western Reservoir. The city of Broomfield built the Broomfield Diversion Ditch to divert Walnut Creek around the reservoir.

Kinner Ditch and Reservoir County Ditch divert water from Coal Creek and deliver it to Woman Creek and eventually to Standley Lake. Last Chance Ditch flows south of RFP and supplies water to Rocky Flats Lake and Twin Lakes. Smart Ditch diverts water from Rocky Flats Lake and transports it offsite to the east. The South Boulder Diversion Canal, located immediately west of the western RFP boundary, diverts water from South Boulder Creek and delivers it to Ralston Reservoir which is a water supply for the city of Denver.

SAMPLE COLLECTION

Eighty-three water samples and 24 sediment samples were collected in FY-93 in support of this project. Over 800 separate chemical analyses were performed on these samples.

Water samples were stabilized in the field with nitric acid using established protocols. Water samples were collected monthly from Ponds A-4, B-5 and C-2 as well as the effluent from STP. Ponds A-1, A-2, A-3, B-1, B-2, B-3, B-4 and C-1 were sampled quarterly. Other locations were sampled a single time. Sediment samples were collected from each of the ponds. A series of sediment samples was collected from the South Interceptor Ditch. Additional samples were collected at locations that may be useful for determining movement of radioactivity at RFP.

EXPERIMENTAL

This project required the analytical measurement of the total alpha activity (gross alpha activity) in the samples, the determination of the amounts of radium, thorium, uranium, plutonium, and americium. These elements represent the major alpha emitting nuclides suspected in the water samples. Below is a brief description of the analytical

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methods used to make the measurements for this study. They are not given in great detail since they have been used in our previous study or they are standard methods of analysis.[3]

Gross Alpha Activity

The total alpha activity was measured according to procedures developed by the Environmental Protection Agency.[4,5] A precisely measured amount of water was evaporated onto a planchet, flamed and counted for gross alpha activity. The count rates were converted to picocuries per liter (pCi/L).

Gross Beta Activity

The gross beta activity was measured according to procedures developed by the Environmental Protection Agency.[4,5] A precisely measured amount of water was evaporated onto a planchet, flamed and counted for gross beta activity. The count rates were converted to picocuries per liter (pCi/L).

Radium

The determination of radium in water samples included the following steps. Radium was co-precipitated onto barium sulfate from a given quantity of water. The barium (radium) sulfate was mounted and counted for alpha particles. The procedure used in this study was developed by the Environmental Protection Agency.[4]

Thorium

Thorium concentrations were determined by inductively coupled plasma mass spectrometry (ICP-MS). The usual protocols established for elemental analysis were followed for the determination of this element. The isotope determined for this analysis is ^{232}Th that is naturally occurring. A de-ionized water sample was run in replicate and produced a thorium signal equivalent to ~0.003 pCi/L. This value was subtracted from each sample.

Uranium

Uranium was determined by thermal ionization mass spectrometry (TIMS). This procedure has been developed in our laboratory and is described in detail in our previous report.[3] This procedure allows the elucidation of the isotopic composition of the uranium in the sample. The following uranium isotopes are determined: ^{234}U, ^{235}U, ^{236}U, and ^{238}U. The uranium isotope ratios can be used to separate the uranium into its naturally occurring and its anthropogenically produced components.

Plutonium

Plutonium was determined by TIMS. This procedure has been developed in our laboratory and is described in detail in our previous report.[3] The procedure allows for the quantification of the isotopic composition of the plutonium isotopes in the water sample, i.e., ^{239}Pu and ^{240}Pu. Measurement of the $^{240}Pu/^{239}Pu$ atom ratio in samples can be used to separate the global fallout component from the RFP component.

Americium

Americium-241 was analyzed by alpha pulse height analysis. The procedure is a standard procedure used in our laboratory. It has been described in detail in our previous report.[3] The ^{241}Am results are reported in Table II. Two liter water samples were traced with ^{243}Am. Samples were quantified by alpha pulse height analyses techniques. Count lengths were varied according to the activity of ^{241}Am present in the sample. A 10,000 minute count length achieves the 0.003 pCi/L limit of detection required by this study. Values reported in Table II that are below 0.003 pCi/L are indistinguishable from detector background.

CONVERSION FROM ATOMS TO ACTIVITY

Thorium, uranium and plutonium were quantified by mass spectrometric measurement techniques. Mass spectrometers measure the number of atoms in a sample. The numbers of atoms are converted to activities by means of the relationship:

$$A = N \lambda$$

A is the activity expressed in disintegrations per unit time. N is the number of atoms of the particular radionuclide and λ is the decay constant that is equal to $0.693/t_{1/2}$ ($t_{1/2}$ is the half-life of the radionuclide). Where there is more than one isotope for the element contributing to the total activity, the total activity of the contributing isotopes is determined by the summation of the individual components. For example, the total uranium activity (A_{total}) is expressed as follows:

$$A_{total} = A_{238U} + A_{236U} + A_{235U} + A_{234U}$$

where A_{238U}, A_{236U}, A_{235U} and A_{234U} are the activity contributions for ^{238}U, ^{236}U, ^{235}U, and ^{234}U.

The half-lives for the radionuclides used for the calculations are given in Table I.

Table I.

Half-lives of Radionuclides Used for Calculating Activities from Atoms Present in the Sample

Radionuclide	Half-Life (years)
Thorium-232	1.400×10^{10}
Uranium-234	2.450×10^{5}
Uranium-235	7.040×10^{8}
Uranium-236	2.342×10^{7}
Uranium-238	4.468×10^{9}
Plutonium-239	2.411×10^{3}
Plutonium-240	6.560×10^{3}

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RESULTS AND DISCUSSION

SURFACE WATERS COLLECTED FROM HOLDING PONDS

Alpha Activity

Table II summarizes the alpha activities measured in surface-waters at RFP. The gross alpha activity and the radium concentrations were obtained using procedures developed by the Environmental Protection Agency (EPA).[4,5] Thorium was measured by ICP-MS. Uranium and plutonium were measured by TIMS. Americium was measured by alpha pulse height analyses techniques.

Table II.

Alpha Activities Measured In Surface-Waters at RFP

Date Sampled	Pond	Sample Number	Gross Alpha* pCi/L	Radium* pCi/L	Thorium† pCi/L	Uranium pCi/L	Plutonium pCi/L	Americium* pCi/L
11/25/92	A1	'SW60293WC						
02/24/93	A1	SW60312WC	1.0	-0.3	0.006	0.73	0.0571	0.026
05/12/93	A1	SW60017JE	2.6	1.0	0.000	2.15	0.0133	0.004
08/24/93	A1	SW60051JE	12.0	0.7	0.000	15.79	0.0261	0.007
11/25/92	A2	SW60294WC	9.2	2.1	0.679	7.25	0.6572	0.162
02/24/93	A2	SW60311WC	6.3	2.9	0.017	3.50	0.0257	0.009
05/12/93	A2	SW60016JE	9.1	3.2	-----	5.59	0.0032	-0.002
08/24/93	A2	SW60052JE	6.1	1.5	0.000	6.41	0.0059	0.001
11/25/92	A3	SW60295WC	8.5	3.9	0.007	5.14	0.0251	0.006
02/24/93	A3	SW60310WC	5.9	3.7	0.001	4.63	0.0030	0.000
05/12/93	A3	SW60015JE	8.5	2.3	0.001	4.48	0.0037	-0.001
08/24/93	A3	SW60053JE	4.6	1.1	-----	4.54	0.0195	0.003

'Sample SW60293WC leaked in transit.
*Where there are negative numbers for gross activity, radium or americium values, the sample count rate was less than the background count rate.
†No value reported for thorium determination indicates that the ICP-MS signal was less than the blank signal for thorium.

Table II. continued

Alpha Activities Measured In Surface-Waters at RFP

Date Sampled	Pond	Sample Number	Gross Alpha* pCi/L	Radium* pCi/L	Thorium† pCi/L	Uranium pCi/L	Plutonium pCi/L	Americium* pCi/L
10/23/92	A4	SW60273WC	3.0	1.2	0.002	1.55	0.0016	0.002
11/24/92	A4	SW60289WC	4.0	0.8	0.004	0.10	0.0061	0.002
12/14/92	A4	SW60301WC	2.3	-1.0	0.029	1.25	0.0091	0.002
01/14/93	A4	SW60305WC	4.5	3.2	0.001	1.50	0.0022	0.000
02/23/93	A4	SW60309WC	7.0	4.1	0.003	3.33	0.0010	-0.001
03/17/93	A4	SW60000JE	4.7	2.2	0.012	2.21	0.0025	0.009
04/21/93	A4	SW60010JE	6.3	3.2	0.012	2.42	0.0036	-0.002
05/12/93	A4	SW60014JE	5.5	2.7	0.002	2.53	0.0014	0.000
06/15/93	A4	SW60027JE	8.0	4.1	0.004	3.53	0.0026	-0.001
07/15/93	A4	SW60031JE	1.5	1.0	0.001	1.63	0.0014	0.000
08/24/93	A4	SW60054JE	1.1	0.7	-----	0.90	0.0179	0.001
02/24/92	B1	SW60317WC	6.3	3.2	0.021	2.87	0.3130	0.104
11/25/92	B1	SW60296WC	3.4	-0.6	0.006	0.70	2.9334	0.851
05/17/93	B1	SW60023JE	5.1	4.0	0.008	3.40	0.1175	0.053
08/24/93	B1	SW60048JE	7.3	2.9	0.000	3.80	0.6855	0.017
11/25/92	B2	SW60297WC	12.9	2.4	0.066	6.95	3.0902	1.030
02/24/93	B2	SW60316WC	7.0	3.1	0.001	4.79	0.0190	0.007
05/17/93	B2	SW60022JE	14.1	8.8	-----	10.97	0.2180	0.081
08/24/93	B2	SW60047JE	7.3	1.9	0.001	5.36	0.3526	0.010
11/25/92	B3	SW60298WC	0.0	-0.3	0.000	0.10	0.1582	0.005
02/24/93	B3	SW60315WC	1.5	0.6	0.001	0.47	0.1020	0.006
05/13/93	B3	SW60021JE	0.6	1.0	-----	0.32	0.0144	0.003
08/24/93	B3	SW60046JE	0.2	-0.2	0.000	0.20	0.2285	0.065
02/24/93	B4	SW60314WC	2.0	0.9	0.001	1.14	0.0777	0.010
05/13/93	B4	SW60020JE	-1.2	-0.9	0.000	1.46	0.0352	0.015
08/24/93	B4	SW60045JE	0.1	0.0	-----	0.20	0.0465	0.009

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Table II. continued

Alpha Activities Measured In Surface-Waters at RFP

Date Sampled	Pond	Sample Number	Gross Alpha* pCi/L	Radium* pCi/L	Thorium† pCi/L	Uranium pCi/L	Plutonium pCi/L	Americium* pCi/L
10/23/92	B5	SW60274WC	-0.6	0.5	0.007	0.65	0.0055	0.000
11/24/92	B5	SW60290WC	-1.5	-0.5	0.003	0.67	0.0111	0.002
12/14/92	B5	SW60302WC	1.3	0.2	0.006	0.66	0.0062	0.003
01/14/93	B5	SW60306WC	4.1	2.1	0.001	0.78	0.0078	0.001
02/23/93	B5	SW60313WC	1.8	0.8	0.003	0.93	0.0058	0.003
03/17/93	B5	SW60001JE	3.2	-0.3	0.007	1.05	0.0043	0.000
04/21/93	B5	SW60011JE	5.2	3.2	0.001	1.75	0.0101	0.002
05/13/93	B5	SW60019JE	-3.0	2.4	-----	1.08	0.0022	-0.002
06/15/93	B5	SW60028JE	2.2	1.5	-----	0.41	0.0018	0.000
07/15/93	B5	SW60032JE	0.4	0.3	0.000	0.65	0.0033	0.000
08/23/93	B5	SW60044JE	1.1	0.0	-----	0.59	0.0057	-0.001
12/13/92	C1	SW60300WC	5.0	1.5	0.001	1.59	0.0049	0.002
02/23/93	C1	SW60320WC	4.9	2.3	0.003	1.85	0.0084	-0.003
05/13/93	C1	SW60025JE	2.6	1.9	0.002	1.48	0.0073	0.002
08/23/93	C1	SW60050JE	2.5	1.0	0.000	1.96	0.0566	0.009
10/21/92	C2	SW60271WC	5.1	-0.7	0.003	1.90	0.0547	0.019
11/25/92	C2	SW60291WC	4.0	1.8	0.004	1.98	0.0270	0.007
12/14/92	C2	SW60303WC	5.3	4.9	0.002	2.12	0.0241	0.006
01/14/93	C2	SW60307WC	4.7	3.5	0.001	2.37	0.0142	0.004
02/23/93	C2	SW60319WC	3.0	1.9	0.002	1.43	0.0043	0.001
03/17/93	C2	SW60002JE	5.1	2.3	0.004	1.23	0.0204	0.005
04/15/93	C2	SW60012JE	4.9	2.0	0.002	2.68	0.0181	0.004
05/13/93	C2	SW60024JE	6.5	2.7	0.002	3.20	0.0305	0.011
06/15/93	C2	SW60029JE	4.9	2.2	0.002	1.96	0.0403	0.015
07/15/93	C2	SW60033JE	5.5	3.7	0.001	2.91	0.0289	0.007
08/23/93	C2	SW60049JE	4.1	1.5	-----	2.50	0.0968	0.035
10/23/92	STP	SW60275WC	-0.9	0.1	0.004	0.02	0.0006	0.000
11/23/92	STP	SW60292WC	0.6	0.5	0.004	0.05	0.0006	0.000
12/14/92	STP	SW60304WC	-1.0	0.5	0.002	0.05	0.0013	-0.001
01/14/93	STP	SW60308WC	-0.1	0.3	0.001	0.31	0.0007	-0.001
02/23/93	STP	SW60318WC	1.0	-0.1	0.000	0.48	0.0005	-0.002
03/17/93	STP	SW60003JE	1.3	0.3	0.001	0.74	0.0008	0.004
04/21/93	STP	SW60013JE	1.8	-0.4	0.000	1.02	0.0021	0.000
05/13/93	STP	SW60018JE	1.1	1.0	-----	0.50	0.0006	0.001
06/15/93	STP	SW60030JE	-0.3	-0.4	0.000	0.91	0.0013	-0.002
07/15/93	STP	SW60034JE	1.2	0.7	0.000	0.54	0.0011	0.000
08/23/93	STP	SW60043JE	0.0	0.0	-----	0.34	0.0013	0.000

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Gross Alpha Activity

The majority of the gross alpha activity detected in waters collected from Ponds A-1, A-2, A-3, B-1 and B-2 was the result of activities at RFP. The largest contributor to the alpha activity measured in these ponds was depleted uranium and plutonium. As a first estimation, the alpha activity measured in Pond C-1 can be attributed to naturally occurring uranium and its daughters. The other ponds contain mixtures of naturally occurring and anthropogenic alpha activities. The gross alpha activity is a measure of all the alpha particle contributions from all the contributing alpha emitters. These include the naturally occurring ^{235}U series, ^{238}U series and ^{232}Th. (Appendix 1 lists the complete decay series.) If members of the series are in equilibrium with the parent nuclides of the series, i.e., ^{235}U, ^{238}U and ^{232}Th, then the total alpha activity attributable to naturally occurring uranium and thorium can be estimated by n times the number of alpha emitters for each series. For ^{235}U n is 7; for ^{238}U n is 8; and for ^{232}Th n is 6. This relationship establishes the upper limit of alpha activity in a sample that can be attributed to naturally occurring radioactivity for samples where secular equilibrium is maintained. Measurement of the gross alpha activity in waters collected at RFP is a crude screening tool. Gross alpha activity measurements may under estimate or over estimate the alpha activity levels in water samples. The largest sample aliquot that should be counted for gross alpha activity is an aliquot size that gives a solid density thickness of 5 mg/cm^2 on the counting planchet. Surface waters collected at RFP contain significant amounts of dissolved salts. In addition, the salts deposited upon evaporation are hydrophilic. These hydrophilic salts must be converted into stable compounds by flaming before alpha counting. Flaming large amounts of residue produces a non-uniform deposit that may attenuate a portion of the alpha activity and cause the activity levels to be under estimated. Anomalously high total alpha activity levels may also be measured. Uranium and radium form water soluble species. Thorium, plutonium, americium and the short-lived naturally occurring uranium and thorium decay products polonium and bismuth are insoluble and tend to concentrate in the sediments. Incorporation of excessive amounts of sediments may lead to over estimates of the alpha activity in the water samples. Therefore, the total alpha activity levels reported must be interpreted in light of the consequence of the built-in problems associated with gross alpha measurements.

Radium

The isotope measured here is radium-226, a member of the ^{238}U decay series. Radium-226 has a half-life approximately 1600 years. It is produced from the decay of ^{230}Th ($t_{1/2}$ = 80,000 years). It produces ^{222}Rn as its decay product. Two other radium isotopes may also be present. These are ^{223}Ra ($t_{1/2}$ = 11.1 days) which is in the natural ^{235}U series and ^{224}Ra ($t_{1/2}$ = 3.64 days) from the natural ^{232}Th series. Depending upon the separation time of the radium from the water and the count time, the degree of equilibrium between the radium isotopes and the parents, and the amounts of each of the three natural series, the radium values may be highly variable. We do not anticipate that ^{223}Ra and ^{224}Ra make a significant contribution to the total radium activity. This, however, does not mean that they are not without value in evaluating the system for alpha activity. These numbers provide an excellent opportunity to estimate the amount of natural alpha activity in the system. Radium is soluble in water and should be present in any natural water system that comes in contact with rock or sediment. All rocks and sediments contain small quantities of naturally occurring uranium and thorium. Consequently, this is a good indicator of the natural component of the alpha activity. There is enough naturally occurring uranium in the water to account for the radium observed in these RFP waters. There is 70-450 times more alpha activity resulting from the decay of naturally occurring radium than alpha activity resulting from the plutonium in the terminal Ponds A-4, B-5 and C-2.

Thorium

It is apparent from the results in Table II that thorium itself does not contribute significantly to the total alpha activity in the RFP waters. This may be explained by the fact that the solubility of thorium in most natural waters is very low. The two most important members of the decay series that may be soluble are radium and radon. (See Appendix 1 for members of the ^{232}Th series.) The range of ^{232}Th concentrations is below detection limit of 0.003 pCi/L to 0.679 pCi/L. The water sample SW60294WC containing 0.679 pCi ^{232}Th/L contained gram quantities of sediments. This sample also contained excessive amounts of uranium and plutonium. The thorium, uranium and plutonium in this sample are most probably associated with the sediment material.

Uranium

Uranium concentrations measured in the waters collected from the holding ponds at RFP ranged from 0.10 pCi/L to 15.79 pCi/L. Uranium measurements were obtained by the isotope dilution technique using TIMS. The measurement technique quantifies the ^{234}U, ^{235}U, ^{236}U and ^{238}U content of the water samples. These data are summarized in Table III.

Table III.

Activity of Specific Uranium Isotopes in Waters

Date Sampled	Pond	Sample Number	Total Uranium pCi/L	U-234 pCi/L	U-235 pCi/L	U-236 pCi/L	U-238 pCi/L
02/29/93	A1	SW60312WC	0.7261	0.2991	0.0186	0.0015	0.4065
05/12/93	A1	SW60017JE	2.1459	0.9466	0.0409	0.0040	1.1544
08/24/93	A1	SW60051JE	15.7994	7.5862	0.2202	0.0309	7.9621
11/25/92	A2	SW60294WC	7.2516	2.8864	0.1310	0.0151	4.2191
02/24/93	A2	SW60311WC	3.4985	1.4472	0.0702	0.0070	1.9741
05/12/93	A2	SW60016JE	5.5906	2.4778	0.1080	0.0105	2.9943
08/24/93	A2	SW60052JE	6.4135	2.7534	0.1163	0.0121	3.5317
11/25/92	A3	SW60295WC	5.1449	2.0257	0.0854	0.0096	3.0242
02/29/93	A3	SW60310WC	4.6302	1.9632	0.0883	0.0080	2.5707
05/12/93	A3	SW60015JE	4.4820	1.8887	0.0783	0.0080	2.5070
08/24/93	A3	SW60053JE	4.5453	1.8590	0.0795	0.0090	2.5978
10/23/92	A4	SW60273WC	1.5500	0.6952	0.0283	0.0018	0.8247
11/25/92	A4	SW60289WC	0.1025	0.0263	0.0031	0.0000	0.0731
12/14/92	A4	SW60301WC	1.2528	0.5647	0.0245	0.0015	0.6621
01/14/93	A4	SW60305WC	1.4984	0.7352	0.0273	0.0016	0.7343
02/29/93	A4	SW60309WC	3.3329	1.5179	0.0613	0.0048	1.7489
03/17/93	A4	SW60000JE	2.2050	0.9906	0.0405	0.0030	1.1708
04/21/93	A4	SW60010JE	2.4169	1.1225	0.0445	0.0029	1.2470
05/12/93	A4	SW60014JE	2.5389	1.2139	0.0464	0.0029	1.2757
06/15/93	A4	SW60027JE	3.5302	1.6457	0.0631	0.0061	1.8153
07/15/93	A4	SW60031JE	1.6257	0.7190	0.0302	0.0018	0.8747
08/23/93	A4	SW60054JE	0.9049	0.4353	0.0173	0.0010	0.4513
11/25/92	B1	SW60296WC	0.7041	0.3716	0.0135	0.0003	0.3187
02/23/93	B1	SW60317WC	2.8725	1.5676	0.0525	0.0025	1.2499
05/13/93	B1	SW60023JE	3.4006	1..8045	0.0633	0.0032	1.5296
08/24/93	B1	SW60048JE	3.8021	1.9081	0.0735	0.0049	1.81566

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Table III. continued

Activity of Specific Uranium Isotopes in Waters

Date Sampled	Pond	Sample Number	Total Uranium pCi/L	U-234 pCi/L	U-235 pCi/L	U-236 pCi/L	U-238 pCi/L
11/25/92	B2	SW60297WC	6.9470	3.7691	0.1399	0.0083	3.0297
02/23/93	B2	SW60316WC	4.7905	2.2466	0.0868	0.0071	2.4500
05/13/93	B2	SW60022JE	10.9670	5.0422	0.1965	0.0259	5.7024
08/24/93	B2	SW60047JE	5.3558	2.4584	0.0963	0.0125	2.7886
11/25/92	B3	SW60298WC	0.0444	0.0153	0.0012	0.0001	0.0278
02/24/93	B3	SW60315WC	0.4671	0.2065	0.0101	0.0005	0.2500
05/13/93	B3	SW60021JE	0.3157	0.1561	0.0059	0.0003	0.1534
08/24/93	B3	SW60046JE	0.2017	0.1242	0.0029	0.0001	0.0745
11/25/92	B4	SW60299WC	1.1159	0.5715	0.0215	0.0011	0.5218
02/24/93	B4	SW60314WC	1.4563	0.6745	0.0301	0.0016	0.7501
05/13/93	B4	SW60020JE	0.6027	0.2650	0.0129	0.0007	0.3241
08/24/93	B4	SW60045JE	0.2037	0.0882	0.0046	0.0004	0.1105
10/23/92	B5	SW60274WC	0.6505	0.3021	0.0143	0.0004	0.3337
11/24/92	B5	SW60290WC	0.6686	0.3289	0.0141	0.0003	0.3253
12/14/92	B5	SW60302WC	0.6563	0.3812	0.0138	0.0001	0.3212
01/14/93	B5	SW60306WC	0.7808	0.3944	0.0152	0.0006	0.3706
02/24/93	B5	SW60313WC	0.9318	0.4661	0.0186	0.0004	0.4467
03/17/93	B5	SW60001JE	1.0507	0.5335	0.0209	0.0007	0.4956
04/21/93	B5	SW60011JE	1.7511	0.8914	0.0346	0.0011	0.8240
05/13/93	B5	SW60019JE	1.0824	0.5451	0.0217	0.0007	0.5149
06/15/93	B5	SW60028JE	0.4086	0.1729	0.0082	0.0003	0.2272
07/15/93	B5	SW60032JE	0.6456	0.3081	0.0136	0.0007	0.3232
08/23/93	B5	SW60044JE	0.5923	0.2786	0.0129	0.0003	0.3005
12/14/92	C1	SW60300WC	1.5887	0.9191	0.0294	0.0000	0.6402
02/23/93	C1	SW60320WC	1.8475	1.0437	0.0352	0.0003	0.7683
05/13/93	C1	SW60025JE	1.4816	0.8336	0.0287	-----	0.6193
08/24/93	C1	SW60050JE	1.9644	1.1230	0.0369	0.0000	0.8045

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Table III. continued

Activity of Specific Uranium Isotopes Waters

Date Sampled	Pond	Sample Number	Total Uranium pCi/L	U-234 pCi/L	U-235 pCi/L	U-236 pCi/L	U-238 pCi/L
10/21/92	C2	SW60271WC	1.9032	0.8550	0.0338	0.0034	1.0110
11/25/92	C2	SW60291WC	1.9759	0.8644	0.0363	0.0042	1.0710
12/14/93	C2	SW60303WC	2.1286	0.9366	0.0407	0.0038	1.1475
01/14/93	C2	SW60307WC	2.3737	1.0856	0.0425	0.0044	1.2412
02/23/93	C2	SW60319WC	1.4309	0.6637	0.0261	0.0025	0.7386
03/17/93	C2	SW60002JE	1.2336	-----	0.0418	-----	1.1918
04/15/93	C2	SW60012JE	2.6754	1.2378	0.0484	0.0043	1.3849
05/13/93	C2	SW60024JE	3.2864	1.5118	0.0596	0.0053	1.7097
06/15/93	C2	SW60029JE	1.9596	0.8713	0.0379	0.0022	1.0482
07/15/93	C2	SW60033JE	2.9120	1.3359	0.0530	0.0048	1.5183
08/24/93	C2	SW60049JE	2.4993	1.1405	0.0456	0.0049	1.3083
10/23/92	STP	SW60275WC	0.0244	0.0126	0.0005	0.0000	0.0113
11/25/92	STP	SW60292WC	0.0477	0.0249	0.0009	0.0001	0.0218
12/14/92	STP	SW60304WC	0.0456	-----	0.0016	-----	0.0440
01/14/93	STP	SW60308WC	0.3076	0.1273	0.0060	0.0005	0.1738
02/23/93	STP	SW60318WC	0.4815	0.2247	0.0097	0.0003	0.2468
03/17/93	STP	SW60003JE	0.7361	0.3575	0.0147	0.0007	0.3632
04/15/93	STP	SW60013JE	1.0177	0.4757	0.0204	0.0010	0.5206
05/13/93	STP	SW60018JE	0.5001	0.2217	0.0102	0.0006	0.2692
06/15/93	STP	SW60030JE	0.9107	0.4654	0.0178	0.0006	0.4269
07/15/93	STP	SW60034JE	0.5452	0.2548	0.0102	0.0005	0.2797
08/24/93	STP	SW60043JE	0.4329	0.1736	0.0066	0.0004	0.1623

The total uranium concentrations reported in Table III is the sum of the ^{234}U, ^{235}U, ^{236}U and ^{238}U activities determined by TIMS. The uranium activities measured in the RFP surface-water samples are higher than the uranium activities measured in some of the reservoirs in the vicinity. The uranium activity levels in the surface-waters collected at RFP are lower than the uranium activities in some wells and springs in the western United States. Table IV reports the highest uranium concentration measured in the RFP ponds during this study. The uranium concentrations in the local reservoirs are the mean uranium concentrations measured by RFP in 1988.[6] The values reported for the selected wells and springs in the western United States were single measurements.[7-15]

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Table IV.

Uranium Concentrations in Waters

Location	Description	Uranium content pCi/L
Pond A-1	RFP Holding Pond	15.79
Pond A-2	RFP Holding Pond	7.25
Pond A-3	RFP Holding Pond	5.14
Pond A-4	RFP Holding Pond	3.53
Pond B-1	RFP Holding Pond	3.80
Pond B-2	RFP Holding Pond	10.97
Pond B-3	RFP Holding Pond	0.47
Pond B-4	RFP Holding Pond	1.46
Pond B-5	RFP Holding Pond	1.75
Pond C-1	RFP Holding Pond	1.96
Pond C-2	RFP Holding Pond	3.20
Boulder	Reservoir	0.4
Dillon	Reservoir	0.5
Great Western	Reservoir	2.05
Ralston	Reservoir	0.9
Standley	Reservoir	1.8
*Shiprock	Well	70
*Gallup	Well	328
*Santa Fe	Well	107
*Casper	Spring	881
*Craig	Well	176
*La Junta	Well	497
*Pueblo	Well	168

* The wells and spring listed in Table IV do not serve as public water supplies for the towns listed in the table.

The atom ratio of ^{238}U to ^{235}U in naturally occurring uranium is always constant. The $^{238}U/^{235}U$ atom ratio in naturally occurring uranium is 137.8 The ^{234}U content of naturally occurring uranium may vary slightly in nature. Uranium-234 is a decay product of ^{238}U and in undisturbed samples the ^{234}U and ^{238}U are in secular equilibrium. Uranium-238 alpha decays to form ^{234}Th. Thorium-234 beta decays with a 24.1 day half-life to form ^{234}Pa. Protactinium-234 beta decays

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with a 6.7 hour half-life to form ^{234}U. Uranium, thorium and protactinium are chemically different and may be separated by natural processes. In addition, the alpha decay of ^{238}U may physically damage the mineral containing the uranium. Physical damage of the mineral's crystal lattice may allow water and air containing carbon dioxide to more readily attack the mineral and increase the availability of the ^{234}U for dissolution. Therefore naturally occurring uranium may be slightly depleted or slightly enriched in ^{234}U relative to ^{238}U. For example, sea water is 15% enriched in ^{234}U relative to ^{238}U. Uranium-236 does not exist in nature. Uranium-236 is produced by neutron capture on ^{235}U in nuclear reactors.

The presence of ^{236}U in the surface-water samples collected at RFP and the variable $^{238}U/^{235}U$ atom ratios detected in water samples collected from the holding ponds prove that anthropogenic uranium is present. Table V summarizes the atom percent of ^{234}U, ^{235}U, ^{236}U and ^{238}U present in each surface-water sample collected at RFP. For comparison the atom percentages of ^{234}U, ^{235}U and ^{238}U in naturally occurring uranium having its ^{234}U and ^{238}U in secular equilibrium are also reported. A naturally occurring uranium sample measured on the TIMS used in this study had a $^{238}U/^{235}U$ atom ratio of 137.80 $\pm$ 0.34. Sample SW60051JE measured on the same instrument had a $^{238}U/^{235}U$ atom ratio of 229.45 $\pm$ 2.29. These values expressed in terms of atom percents ^{235}U are (0.7204 $\pm$ 0.0018) % ^{235}U for the naturally occurring uranium and (0.4339 $\pm$ 0.0043) % ^{235}U for sample SW60051JE.

Table V.

Atom Percent of Uranium Isotopes Present in RFP Surface-Waters

Date Sampled	Pond	Sample Number	% U-234	% U-235	% U-236	% U-238
Natural Uranium			0.0057	0.7204	0.0000	99.2739
02/29/93	A1	SW60312WC	0.0040	0.7138	0.0020	99.2802
05/12/93	A1	SW60017JE	0.0045	0.5549	0.0018	99.4388
08/24/93	A1	SW60051JE	0.0052	0.4339	0.0020	99.5589
11/25/92	A2	SW60294WC	0.0037	0.4869	0.0019	99.5075
02/24/93	A2	SW60311WC	0.0040	0.5568	0.0018	99.4373
05/12/93	A2	SW60016JE	0.0045	0.5652	0.0018	99.4284
08/24/93	A2	SW60052JE	0.0043	0.4259	0.0018	99.4777

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Table V. continued

Atom Percent of Uranium Isotopes Present in RFP Surface-Waters

Date Sampled	Pond	Sample Number	% U-234	% U-235	% U-236	% U-238
Natural Uranium			0.0057	0.7204	0.0000	99.2739
11/25/92	A3	SW60295WC	0.0037	0.4427	0.0017	99.5520
02/29/93	A3	SW60310WC	0.0042	0.5385	0.0016	99.4557
05/12/93	A3	SW60015JE	0.0041	0.4897	0.0017	99.5045
08/24/93	A3	SW60053JE	0.0039	0.4801	0.0018	99.5142
10/23/92	A4	SW60273WC	0.0046	0.5377	0.0011	99.4566
11/24/92	A4	SW60289WC	0.0020	0.6604	0.0003	99.3373
12/14/92	A4	SW60301WC	0.0046	0.5798	0.0011	99.4144
01/14/93	A4	SW60305WC	0.0055	0.5827	0.0012	99.4107
02/23/93	A4	SW60309WC	0.0047	0.5490	0.0014	99.4448
03/17/93	A4	SW60000JE	0.0046	0.5425	0.0014	99.4516
04/21/93	A4	SW60010JE	0.0049	0.5588	0.0012	99.4351
05/12/93	A4	SW60014JE	0.0052	0.5693	0.0012	99.4243
06/15/93	A4	SW60027JE	0.0049	0.5445	0.0017	99.4488
07/15/93	A4	SW60031JE	0.0045	0.5417	0.0011	99.4527
08/23/93	A4	SW60054JE	0.0053	0.6001	0.0012	99.3935
11/25/92	B1	SW60296WC	0.0064	0.6649	0.0006	99.3282
02/24/93	B1	SW60317WC	0.0068	0.6573	0.0010	99.3348
05/13/93	B1	SW60023JE	0.0064	0.6473	0.0011	99.3451
08/24/93	B1	SW60048JE	0.0059	0.6340	0.0014	99.3588
11/25/92	B2	SW60297WC	0.0068	0.7225	0.0014	99.2693
02/23/92	B2	SW60316WC	0.0050	0.5553	0.0015	99.4382
05/13/93	B2	SW60022JE	0.0048	0.5400	0.0024	99.4528
08/24/93	B2	SW60047JE	0.0048	0.5414	0.0023	99.4514
11/25/92	B3	SW60298WC	0.0030	0.6737	0.0017	99.3216
02/24/93	B3	SW60315WC	0.0045	0.6349	0.0011	99.3595
05/13/93	B3	SW60021JE	0.0055	0.6018	0.0010	99.3916
08/24/93	B3	SW60046JE	0.0091	0.6143	0.0009	99.3757
11/25/92	B4	SW60299WC	0.0060	0. 6462	0.0011	99.3468
02/24/93	B4	SW60314WC	0.0049	0.6290	0.0011	99.3650
05/13/93	B4	SW60020JE	0.0045	0.6229	0.0011	99.3715
08/24/93	B4	SW60045JE	0.0043	0.6480	0.0018	99.3458

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Table V. continued

Atom Percent of Uranium Isotopes Present in RFP Surface-Waters

Date Sampled	Pond	Sample Number	% U-234	% U-235	% U-236	% U-238
Natural Uranium			0.0057	0.7204	0.0000	99.2739
10/23/93	B5	SW60274WC	0.0049	0.6716	0.0006	99.3229
11/24/92	B5	SW60290WC	0.0056	0.6788	0.0005	99.3153
12/14/92	B5	SW60302WC	0.0065	0.6721	0.0001	99.3213
01/14/93	B5	SW60306WC	0.0058	0.6413	0.0009	99.3521
02/23/93	B5	SW60313WC	0.0057	0.6508	0.0005	99.3341
03/17/93	B5	SW60001JE	0.0059	0.6615	0.0007	99.3319
04/21/93	B5	SW60011JE	0.0059	0.6568	0.0007	99.3366
05/13/93	B5	SW60019JE	0.0058	0.6593	0.0007	99.3342
06/15/93	B5	SW60028JE	0.0042	0.5679	0.0008	99.4272
07/15/93	B5	SW60032JE	0.0052	0.6569	0.0011	99.3368
08/23/93	B5	SW60044JE	0.0050	0.6728	0.0006	99.3216
12/14/92	C1	SW60300WC	0.0078	0.7175	0.0000	99.2747
02/24/93	C1	SW60320WC	0.0074	0.7170	0.0002	99.2754
05/13/93	C1	SW60025JE	0.0073	0.7242	-----	99.2684
08/23/93	C1	SW60050JE	0.0076	0.7176	0.0000	99.2748
10/23/92	C2	SW60271WC	0.0046	0.5242	0.0018	99.4694
11/25/92	C2	SW60291WC	0.0044	0.5308	0.0020	99.4628
12/14/92	C2	SW60303WC	0.0045	0.5552	0.0017	99.4386
01/14/93	C2	SW60307WC	0.0048	0.5370	0.0018	99.4564
02/23/93	C2	SW60319WC	0.0049	0.5534	0.0017	99.4400
03/17/93	C2	SW60002JE	-----	0.5502	-----	99.4498
04/15/93	C2	SW60012JE	0.0049	0.5471	0.0016	99.4464
05/13/93	C2	SW60024JE	0.0048	0.5465	0.0016	99.4471
06/15/93	C2	SW60029JE	0.0045	0.5668	0.0011	99.4276
07/15/93	C2	SW60033JE	0.0048	0.5469	0.0016	99.4466
08/24/93	C2	SW60049JE	0.0048	0.5426	0.0020	99.4471
10/23/92	STP	SW60275WC	0.0060	0.6414	0.0009	99.3516
11/25/92	STP	SW60292WC	0.0062	0.6420	0.0004	99.3515
12/14/92	STP	SW60304WC	-----	0.5811	-----	99.4189
01/14/93	STP	SW60308WC	0.0040	0.5430	0.0014	99.4516
02/23/93	STP	SW60318WC	0.0050	0.6141	0.0007	99.3803
03/17/93	STP	SW60003JE	0.0054	0.6344	0.0010	99.3592
04/15/93	STP	SW60013JE	0.0050	0.6137	0.0010	99.3803
05/13/93	STP	SW60018JE	0.0045	0.5907	0.0012	99.4036
06/15/93	STP	SW60030JE	0.0059	0.6542	0.0008	99.3391
07/15/93	STP	SW60034JE	0.0050	0.5723	0.0009	99.4218
08/24/93	STP	SW60043JE	0.0058	0.6390	0.0012	99.3539

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Naturally occurring uranium contains 0.7204 atom percent ^{235}U. Most of the surface-water samples measured at RFP were depleted in ^{235}U. The amount of ^{235}U present in depleted uranium varies. The majority of depleted uranium produced in this country contains 0.2% ^{235}U to 0.5% ^{235}U.[18] Table VI illustrates the maximum amount of depleted uranium that is present in each water sample collected at RFP. The values derived assume a simple two component system. The data reported in Table VI assume that the depleted uranium released by RFP contains 0.5% ^{235}U. This conservative estimate determines the maximum amount of depleted uranium present in the surface-waters.

Estimates are made using the following relationship:

$$(N238/N235)obs = \{(N238/N235)dep\} \{F\} + \{(N238/N235)nat\} \{1-F\}$$

(N238/N235)obs is the $^{238}U/^{235}U$ atom ratio measured in the sample, (N238/N235)dep is the $^{238}U/^{235}U$ atom ratio in the depleted uranium and (N238/N235)nat is the $^{238}U/^{235}U$ atom ratio in naturally occurring uranium. F is the fraction of depleted uranium in the sample and {1-F} is the fraction of the uranium that is naturally occurring. This equation can be solved to provide an estimate of amount of depleted uranium present in the samples.

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Table VI.

Maximum Percentage of Uranium in Water that is Attributable to Release of Depleted Uranium at RFP

Date Sampled	Pond	Sample Number	Maximum Percentage of Uranium Attributable to Release of Depleted Uranium From RFP
02/29/93	A1	SW60312WC	2
05/12/93	A1	SW60017JE	50
08/24/93	A1	SW60051JE	100
11/25/92	A2	SW60294WC	81
05/12/93	A2	SW60016JE	46
02/29/93	A2	SW60311WC	50
08/24/93	A2	SW60052JE	66
11/25/92	A3	SW60295WC	100
02/29/93	A3	SW60310WC	57
05/12/93	A3	SW60015JE	80
08/24/93	A3	SW60053JE	100
10/23/92	A4	SW60273WC	81
11/24/92	A4	SW60289WC	17
12/14/92	A4	SW60301WC	41
01/14/93	A4	SW60305WC	40
02/23/93	A4	SW60309WC	53
03/17/93	A4	SW60000JE	58
04/21/93	A4	SW60010JE	49
05/12/93	A4	SW60014JE	45
06/15/93	A4	SW60027JE	54
07/15/93	A4	SW60031JE	56
08/23/93	A4	SW60054JE	31
11/25/92	B1	SW60296WC	14
05/13/93	B1	SW60023JE	23
02/24/93	B1	SW60317WC	17
08/24/93	B1	SW60048JE	23
11/25/92	B2	SW60297WC	0
05/13/93	B2	SW60022JE	57
02/24/93	B2	SW60316WC	50
08/24/93	B2	SW60047JE	56
11/25/92	B3	SW60298WC	12
02/24/93	B3	SW60315WC	23
05/13/93	B3	SW60021JE	35
08/24/93	B3	SW60046JE	36

Table VI. continued

Maximum Percentage of Uranium in Water that is Attributable to Release of Depleted Uranium at RFP

Date Sampled	Pond	Sample Number	Maximum Percentage of Uranium Attributable to Release of Depleted Uranium From RFP
11/25/92	B4	SW60299WC	19
05/13/93	B4	SW60020JE	26
02/29/93	B4	SW60314WC	25
08/24/93	B4	SW60045JE	19
10/23/92	B5	SW60274WC	13
11/24/92	B5	SW60290WC	10
12/14/92	B5	SW60302WC	12
01/14/93	B5	SW60306WC	21
02/23/93	B5	SW60313WC	18
03/17/93	B5	SW60001JE	18
04/21/93	B5	SW60011JE	17
05/13/93	B5	SW60019JE	16
06/15/93	B5	SW60028JE	46
07/15/93	B5	SW60032JE	16
08/23/93	B5	SW60044JE	12
12/03/92	C1	SW60300WC	1
05/13/93	C1	SW60025JE	0
02/23/93	C1	SW60320WC	1
08/23/93	C1	SW60050JE	0
10/23/92	C2	SW60271WC	63
11/25/92	C2	SW60291WC	60
12/14/92	C2	SW60303WC	51
01/14/93	C2	SW60307WC	58
02/23/93	C2	SW60319WC	51
03/17/93	C2	SW60002JE	52
04/15/93	C2	SW60012JE	53
05/13/93	C2	SW60024JE	53
06/15/93	C2	SW60029JE	46
07/15/93	C2	SW60033JE	51
08/24/93	C2	SW60049JE	54

Table VI. continued

Maximum Percentage of Uranium in Water that is Attributable to Release of Depleted Uranium at RFP

Date Sampled	Pond	Sample Number	Maximum Percentage of Uranium Attributable to Release of Depleted Uranium From RFP
10/23/92	STP	SW60275WC	20
11/23/92	STP	SW60292WC	20
12/14/92	STP	SW60304WC	55
01/14/93	STP	SW60308WC	56
02/23/93	STP	SW60318WC	29
03/17/93	STP	SW60003JE	24
04/15/93	STP	SW60013JE	29
05/13/93	STP	SW60018JE	37
06/15/93	STP	SW60030JE	17
07/15/93	STP	SW60034JE	43
08/24/93	STP	SW60043JE	21

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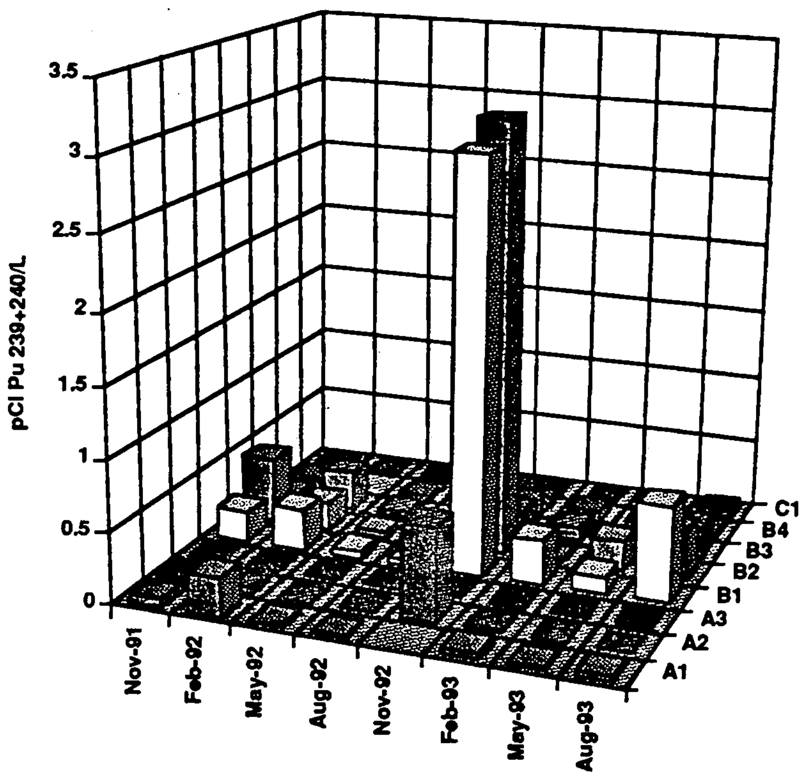
Plutonium

The plutonium content of Ponds A-1, A-2, A-3, B-1, B-2, B-3, B-4 and C-1 was measured quarterly. The results are presented in Figure 3. The data obtained before October 1, 1992 have been previously reported.[3]

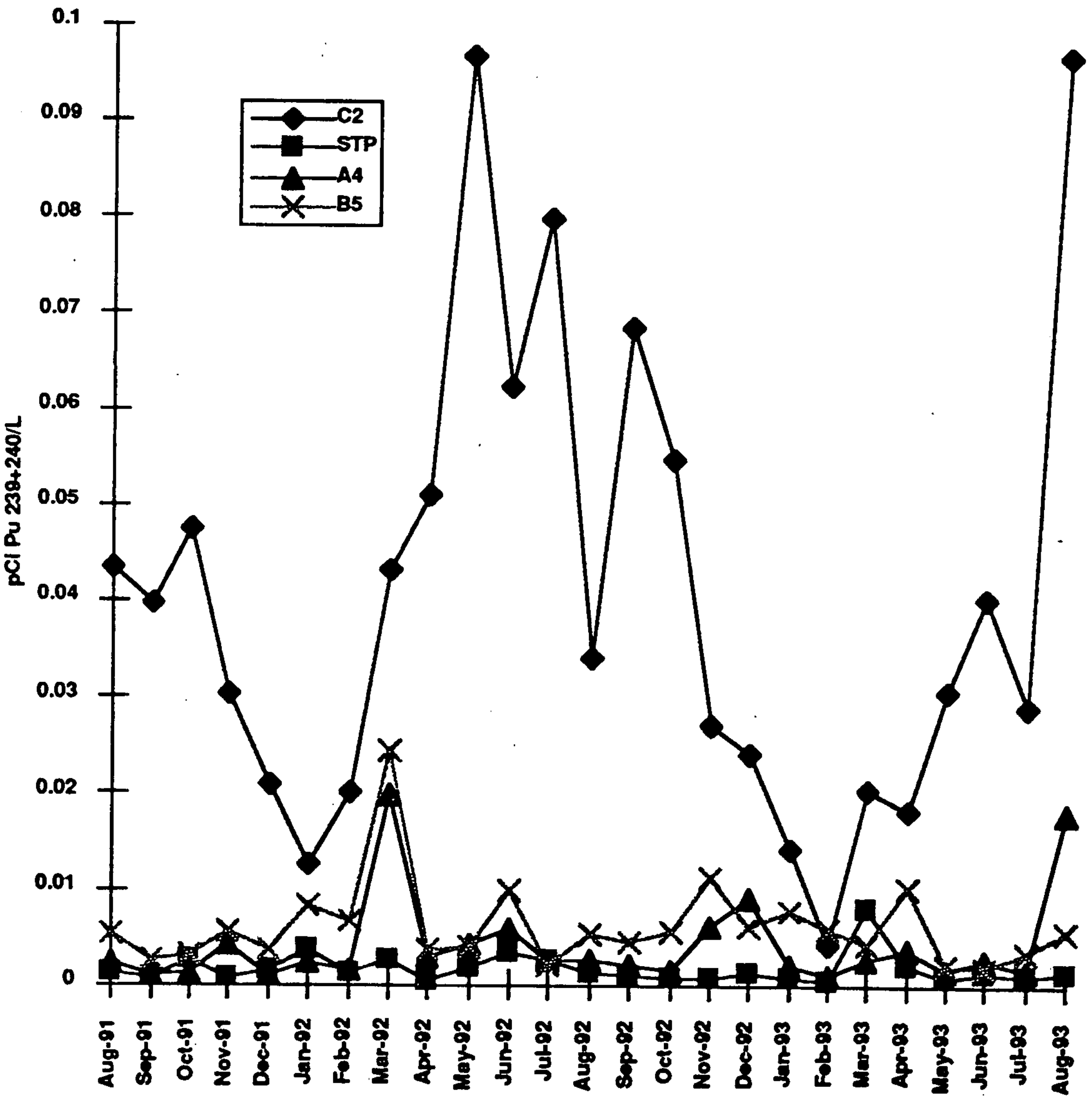
Figure 3.
Plutonium Concentrations In Holding Ponds at RFP that are Analyzed Quarterly



The plutonium concentrations observed in Ponds A-2, B-1 and B-2 during November, 1992 are significantly higher than any other measurements of RFP pond waters. These samples contained significant (multiple grams) quantities of sediments. The sediments in the ponds contain more plutonium on a per gram basis than the waters. Sediments may be re-suspended by wind or they may be incorporated during sampling. Inclusion of sediments in water samples can bias analytical results.

Ponds A-4, B-5, C-2 and the effluent from STP were sampled monthly. The plutonium concentrations in these ponds are presented in Figure 4.

Figure 4
Plutonium Concentrations in Locations Analyzed Monthly



There appears to be a seasonal variation in the plutonium concentrations in Pond C-2. Plutonium concentrations are higher in the

TIMS measures ^{239}Pu and ^{240}Pu separately. The $^{240}Pu/^{239}Pu$ atom ratio can be used to distinguish plutonium originating at RFP from Global Fallout. The Global Fallout deposited in the vicinity of RFP has a $^{240}Pu/^{239}Pu$ atom ratio of 0.169 ± 0.004.[19] The plutonium released by RFP has a $^{240}Pu/^{239}Pu$ atom ratio of 0.06 ± 0.01.[20] Table VII lists the $^{240}Pu/^{239}Pu$ atom ratios measured in surface-waters collected at RFP.

Table VII.

$^{240}Pu/^{239}Pu$ Atom Ratios Measured in RFP Pond Waters

Date Sampled	Sample Number	Pond	$^{240}Pu/^{239}Pu$
02-24-93	SW60312WC	A1	0.054 + 0.004
05-12-93	SW60017JE	A1	0.063 ± 0.009
08-24-93	SW60051JE	A1	0.055 ± 0.005
11-25-92	SW60294WC	A2	0.064 ± 0.007
02-24-93	SW60311WC	A2	0.058 ± 0.010
05-12-93	SW60016JE	A2	0.055 ± 0.008
05-12-93	SW60295WC	A3	0.059 ± 0.005
11-25-92	SW60296WC	B1	0.063 ± 0.001
02-24-93	SW60317WC	B1	0.067 ± 0.004
05-17-93	SW60023JE	B1	0.061 ± 0.002
08-24-93	SW60048JE	B1	0.062 ± 0.005
11-25-92	SW60297WC	B2	0.055 ± 0.002
05-17-93	SW60022JE	B2	0.071 ± 0.006
08-24-93	SW60047JE	B2	0.061 ± 0.005
11-25-92	SW60298WC	B3	0.063 ± 0.005
02-24-93	SW60314WC	B3	0.065 ± 0.003
08-24-93	SW60046JE	B3	0.064 ± 0.005

Table VII. continued

$^{240}Pu/^{239}Pu$ Atom Ratios Measured in RFP Pond Waters

Date Sampled	Sample Number	Pond	$^{240}Pu/^{239}Pu$
11-25-93	SW60299WC	B4	0.060 ± 0.003
02-24-93	SW60313WC	B4	0.066 ± 0.008
05-13-93	SW60020JE	B4	0.059 ± 0.007
08-24-93	SW60045JE	B4	0.060 ± 0.005
02-24-93	SW60320WC	C1	0.064 ± 0.009
05/13/93	SW60025JE	C1	0.069 ± 0.006
08-23-93	SW60050JE	C1	0.057 ± 0.005
01-16-92	SW60200WC	C2	0.054 ± 0.004
11-25-92	SW60291WC	C2	0.052 ± 0.005
12-14-92	SW60303WC	C2	0.065 ± 0.003
01-14-93	SW60307WC	C2	0.062 + 0.005
03-17-93	SW60002JE	C2	0.056 ± 0.009
04-21-93	SW60013JE	C2	0.056 ± 0.001
05-13-93	SW60024JE	C2	0.062 ± 0.002
06-15-93	SW60029JE	C2	0.055 ± 0.007
07-15-93	SW60033JE	C2	0.051 + 0.010

All of the $^{240}Pu/^{239}Pu$ atom ratios reported in Table VII are consistent with the atom ratios of the plutonium released by RFP.

Americium

The americium to plutonium alpha activity ratio in materials processed at RFP is not constant. The alpha activity ratio ranges from 0.2-0.4. This variation is explained by the facts that different batches of plutonium contain different amounts of ^{241}Pu and the geochemical behavior of plutonium and americium are different. The plutonium content of the water samples was always higher than the americium content. All samples that contained measurable amounts of americium also contained measurable amounts of plutonium.

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Gross Beta Activity

Table VIII reports the gross beta activity measured in the surface-water samples. The gross alpha activity, uranium alpha activity and radium alpha activity are also included in Table VIII.

Table VIII.

Gross Beta Activity Detected in Surface-Waters at RFP

Date Sampled	Pond	Sample Number	Gross Alpha pCi/L	Radium pCi/L	Uranium pCi/L	Gross Beta pCi/L
02/29/93	A1	SW60312WC	1.5	-0.3	0.73	-0.9
05/12/93	A1	SW60017JE	1.5	1.0	2.15	6.3
08/24/93	A1	SW60051JE	12	0.7	15.79	5.1
11/25/92	A2	SW60294WC	9.2	2.1	7.25	18.1
02/24/93	A2	SW60311WC	6.3	2.9	3.50	10.6
05/12/93	A2	SW60016JE	9.1	3.2	5.59	3.2
08/24/93	A2	SW60052JE	6.1	1.5	6.41	13.5
11/25/92	A3	SW60295WC	8.5	3.9	5.14	19.1
02/29/93	A3	SW60310WC	5.9	3.7	4.63	11.9
05/12/93	A3	SW60015JE	8.5	2.3	4.48	12.7
08/24/93	A3	SW60053JE	4.6	1.1	4.54	10.2
10/23/92	A4	SW60273WC	3.0	1.2	1.55	3.7
11/25/92	A4	SW60289WC	4.0	0.8	0.10	3.8
12/14/92	A4	SW60301WC	2.3	-1.0	1.25	4.7
01/14/93	A4	SW60305WC	4.5	3.2	1.50	4.2
02/29/93	A4	SW60309WC	7.0	4.1	3.33	6.2
03/17/93	A4	SW60000JE	4.7	2.2	2.25	5.0
04/21/93	A4	SW60010JE	6.3	3.2	2.45	6.2
05/12/93	A4	SW60014JE	5.5	2.7	2.53	7.2
06/15/93	A4	SW60027JE	8.0	4.1	3.53	5.0
07/15/93	A4	SW60031JE	1.5	1.0	1.63	3.2
08/23/93	A4	SW60054JE	1.1	0.7	0.90	4.9
11/25/92	B1	SW60296WC	3.4	-0.6	0.70	5.9
02/23/93	B1	SW60317WC	6.3	3.2	2.87	15.0
05/13/93	B1	SW60023JE	5.1	4.0	3.40	8.7
08/24/93	B1	SW60048JE	7.3	2.9	3.80	9.5

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Table VIII. continued

Gross Beta Activity Detected in Surface-Waters at RFP

Date Sampled	Pond	Sample Number	Gross Alpha pCi/L	Radium pCi/L	Uranium pCi/L	Gross Beta pCi/L
11/25/92	B2	SW60297WC	12.9	2.4	6.95	18.5
02/23/92	B2	SW60316WC	7.0	3.1	4.79	12.9
05/13/93	B2	SW60022JE	14.1	8.8	10.97	20.1
08/24/93	B2	SW60047JE	7.3	1.9	5.36	10.3
11/25/92	B3	SW60298WC	0.0	-0.3	0.10	1.2
02/24/93	B3	SW60315WC	1.5	0.6	0.47	3.7
05/13/93	B3	SW60021JE	0.6	1.0	0.32	5.9
08/24/93	B3	SW60046JE	0.2	-0.2	0.20	4.1
11/25/92	B4	SW60299WC	-0.2	0.8	1.12	9.5
02/24/93	B4	SW60314WC	2.0	0.9	1.46	3.6
05/13/93	B4	SW60020JE	-1.2	-0.9	0.60	-0.1
08/24/94	B4	SW60045JE	0.1	0.0	0.20	4.1
10/23/92	B5	SW60274WC	-0.6	0.5	0.65	4.1
11/24/92	B5	SW60290WC	-1.5	-0.5	0.67	0.0
12/14/92	B5	SW60302WC	1.3	0.2	0.66	6.7
01/14/93	B5	SW60306WC	4.1	2.1	0.78	4.8
02/24/93	B5	SW60313WC	1.8	0.8	0.93	-2.9
03/17/93	B5	SW60001JE	3.2	-0.3	1.09	5.3
04/21/93	B5	SW60011JE	5.2	3.2	1.79	3.7
05/13/93	B5	SW60019JE	-3.0	2.4	1.08	4.2
07/15/93	B5	SW60032JE	0.4	0.3	0.65	1.5
08/23/93	B5	SW60044JE	1.1	0.0	0.59	3.0
12/14/92	C1	SW60300WC	5.0	1.5	1.59	5.0
02/23/93	C1	SW60320WC	4.9	2.3	1.85	5.2
05/13/93	C1	SW60025JE	2.6	1.9	1.48	4.3
08/23/93	C1	SW60050JE	2.5	1.0	1.96	4.9

Table VIII. continued

Gross Beta Activity Detected in Surface-Waters at RFP

Date Sampled	Pond	Sample Number	Gross Alpha pCi/L	Radium pCi/L	Uranium pCi/L	Gross Beta pCi/L
10/21/92	C2	SW60271WC	5.1	-0.7	1.90	3.2
11/25/92	C2	SW60291WC	4.0	1.8	1.98	5.8
12/14/92	C2	SW60303WC	5.3	4.9	2.13	5.0
01/14/93	C2	SW60307WC	4.7	3.5	2.37	2.1
02/23/93	C2	SW60319WC	3.0	1.9	1.43	3.7
03/17/93	C2	SW60002JE	5.1	2.3	2.27	4.2
04/15/93	C2	SW60012JE	4.9	2.0	2.65	5.5
05/13/93	C2	SW60024JE	6.5	2.7	3.20	6.8
06/15/93	C2	SW60029JE	4.9	2.2	1.96	6.0
07/15/93	C2	SW60033JE	5.5	3.7	2.91	4.9
08/24/93	C2	Sw60049JE	4.1	1.5	2.50	6.5
10/23/92	STP	SW60275WC	-0.9	0.1	0.02	2.3
11/25/92	STP	SW60292WC	0.6	0.5	0.05	-1.0
12/14/92	STP	SW60304WC	-1.0	0.5	0.05	1.8
01/14/93	STP	SW60308WC	-0.1	0.3	0.31	1.1
02/23/93	STP	SW60318WC	1.0	-0.1	0.48	3.4.
03/17/93	STP	SW60003JE	1.3	0.3	0.74	0.0
04/15/93	STP	SW60013JE	1.8	-0.4	1.02	-1.3
05/13/93	STP	SW60018JE	1.1	1.0	0.50	2.7
05/15/93	STP	SW60030JE	-0.3	-0.4	0.91	1.5
07/15/93	STP	SW60034JE	1.2	0.7	0.54	-0.9
08/24/93	STP	SW60043JE	0.0	0.0	0.34	-2.1

The gross beta activity is a measure of all the beta particle contributions from all the beta emitters. These include the naturally occurring ^{235}U series, ^{238}U series, ^{232}Th series and ^{40}K. (Appendix 1 lists the complete uranium and thorium decay series.) If members of these series are in radioactive equilibrium with the parent nuclides of the series, i.e., ^{235}U, ^{238}U and ^{232}Th, then the total beta activity attributable to naturally occurring uranium and thorium can be estimated by n times the number of beta emitters for each series. For ^{235}U n is 4; for ^{238}U n is 6; and for ^{232}Th n is 4. The beta activities resulting from the decay of the uranium and thorium series summed with the beta activity associated with the decay of ^{40}K establish the upper limit of beta activity in a sample that can be attributed to naturally occurring radioactivity.

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Measurement of the gross beta activity in waters collected at RFP is a crude screening tool. There is enough naturally occurring uranium and potassium present in the surface-waters at RFP so that the calculated amount of natural beta emitting radionuclides more than account for the gross beta activity measured in the water samples. The ponds may also contain varying amounts of anthropogenic beta activity. This anthropogenic beta activity is indistinguishable from the naturally occurring beta activity and apparently does not seem to have contributed significantly to the total beta activity levels detected in the water samples.

Plutonium-241 is a beta emitter present in plutonium. The beta decay of the ^{241}Pu in RFP plutonium does not contribute significantly to the beta activity in the surface-waters at RFP. For example, the highest plutonium concentration measured in this study was detected in the sediment sample SW60286WC. The sample contained 4.34 pCi ^{238}Pu/g, 152.26 pCi $^{239+240}$Pu/g and 10.8 pCi ^{241}Pu/g. These data imply that the ^{241}Pu/$^{239+240}$Pu activity ratio in RFP plutonium is approximately 0.07. Therefore, for every 1 pCi of alpha activity resulting from the decay of $^{239+240}$Pu there should be 0.07 pCi of beta activity resulting from the decay of the ^{241}Pu. No significant beta activity can be attributed to the decay products of ^{241}Am.

A 90 L aliquot of water collected from Pond C-2 was evaporated to dryness. The resulting salts were pressed into a standard counting geometry and analyzed by gamma ray spectrometric measurement techniques. Most radionuclides that decay by beta emission also produce gamma rays during the decay process. (^{3}H, ^{90}Sr, ^{99}Tc, ^{147}Pm and ^{129}I are notable exceptions.) Cesium-137, ^{40}K and the daughters of ^{238}U were detected in the 90L water sample. Cesium-137 is a beta emitting radionuclide that was deposited as global fallout. Potassium-40 is a naturally occurring beta emitting radionuclide. The ^{137}Cs concentration was measured to be 0.04 pCi/L and the ^{40}K concentration was measured to be 4.5 pCi/L. The ^{137}Cs activity level is consistent with that expected from global fallout and the ^{40}K level is consistent with levels measured in other waters in Colorado and New Mexico.[16,17] No extraneous radio-isotopes were observed.

SEDIMENT AND SOIL SAMPLES

Sediment samples were collected from each of the ponds at RFP. Soil samples were also collected in the South Interceptor Ditch and at other strategic locations believed to affect surface-waters at RFP. Resuspension of sediments can significantly increase the actinide concentrations in waters. Transport of sediments can relocate the actinides. Percolation of water through the sediments can affect ground waters. The sediment samples collected for this study were analyzed for radioactive content. Sampling locations are summarized in Table IX. Actinides concentrations in the sediments are reported in Table X.

Table IX.

Sediment and Soil Sample Collection Sites

Date Sampled	Sample Number	Location	Description
08/12/93	SW60038JE	SW092	A-1 Bypass Sediment
11/06/92	SW60277WC	SED60392	A-1 Pond Sediment
11/12/92	SW60285WC	SED60892	A-2 Pond Sediment
11/18/92	SW60287WC	SED62892	A-2 Pond Sediment
10/21/92	SW60270WC	SED1392	A-3 Pond Sediment
10/19/92	SW60268WC	-----	A-4 Pond Sediment
11/16/92	SW60286WC	SED62392	B-1 Pond Sediment
10/27/92	SW60276WC	SED63392	B-3 Pond Sediment
10/22/92	SW60272WC	SED3897	B-4 Pond Sediment
10/20/92	SW60269WC	SED64392	B-5 Pond Sediment
06/08/93	SW60026JE	SED51493	C-1 Pond Sediment
11/09/92	SW60278WC	C-1 Pond	C-1 Pond Sediment
11/10/92	SW60279WC	SED913	C-2 Pond Sediment
08/12/93	SW60035JE	SW01593	Coal Creek:Woman Creek Headgate
08/12/93	SW60040JE	SW097	Landfill Pond Sediments
03/22/93	SW60004JE	SW054	South Interceptor Ditch
03/22/93	SW60005JE	SW070	South Interceptor Ditch
03/22/93	SW60006JE	SW044	South Interceptor Ditch
03/22/93	SW60007JE	SW044(N)	South Interceptor Ditch
03/22/93	SW60008JE	SW036	South Interceptor Ditch
03/22/93	SW60009JE	SW038	South Interceptor Ditch
09/23/93	SW60059JE	SW038A	South Interceptor Ditch
08/12/93	SW60041JE	SW035	South Interceptor Ditch
11/19/92	SW60288WC	SED64892	Walnut and Indian Sample Pond

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Table X

Actinide Elements Detected in Sediments and Soils

Sample Number	Description	Thorium pCi/g	Uranium pCi/g	Plutonium pCi/g	Americium pCi/g
SW60038JE	A-1 Bypass Sediments	-----	1.86	7.624	2.268
SW60277WC	A-1 Pond Sediment	1.18	4.37	2.375	0.831
SW60285WC	A-2 Pond Sediment	1.24	6.01	2.829	0.720
SW60270WC	A-3 Pond Sediment	2.26	1.67	0.674	0.235
SW60268WC	A-4 Pond Sediment	1.60	1.17	0.105	0.004
SW60286WC	B-1 Pond Sediment	2.11	8.74	152.256	30.909
SW60287WC	B-2 Pond Sediment	1.09	4.16	12.973	2.727
SW60276WC	B-3 Pond Sediment	2.09	6.50	11.335	2.818
SW60272WC	B-4 Pond Sediment	1.67	1.26	1.062	0.250
SW60269WC	B-5 Pond Sediment	1.85	0.97	0.348	0.066
SW60026JE	C-1 Pond Sediment	1.62	0.57	>0.002	-0.002
SW60278WC	C-1 Pond Sediment	1.43	1.95	***LIA	-0.001
SW60279WC	C-2 Pond Sediment	2.30	2.75	2.728	0.682
SW60035JE	*Coal Creek:Woman Creek	-----	0.70	>0.001	0.000
SW60040JE	Landfill Pond Sediments	-----	0.85	0.016	0.002
SW60004JE	South Interceptor Ditch	2.79	1.14	0.548	0.126
SW60005JE	South Interceptor Ditch	2.22	1.00	0.094	0.024
SW60006JE	South Interceptor Ditch	1.52	0.73	0.014	0.003
SW60007JE	South Interceptor Ditch	1.92	0.80	0.016	0.006
SW60008JE	South Interceptor Ditch	1.32	1.60	0.384	0.100
SW60009JE	South Interceptor Ditch	0.68	0.48	0.004	-0.002
SW60059JE	South Interceptor Ditch	-----	1.67	0.007	-----
SW60041JE	South Interceptor Ditch	1.65	2.54	0.158	0.041
SW60288WC	**Walnut and Indian	-----	1.41	0.155	0.005

* Coal Creek: Woman Creek Headgate
** Walnut and Indian Sample Pond
*** Sample Lost in analyses
----- Sample not analyzed for specific isotope

Comparison of the plutonium activities reported in Table II for water samples to the plutonium activities reported in Table X for sediment samples indicates that one gram of sediment from a holding pond contains approximately 50 times more plutonium than 1 liter of water from the pond. The highest plutonium concentrations were detected in sediments collected from Ponds B-1, B-2 and B-3. More plutonium was detected in the sediment collected from the A-1 Bypass than the sediments collected in the A series ponds. The sediment collected from

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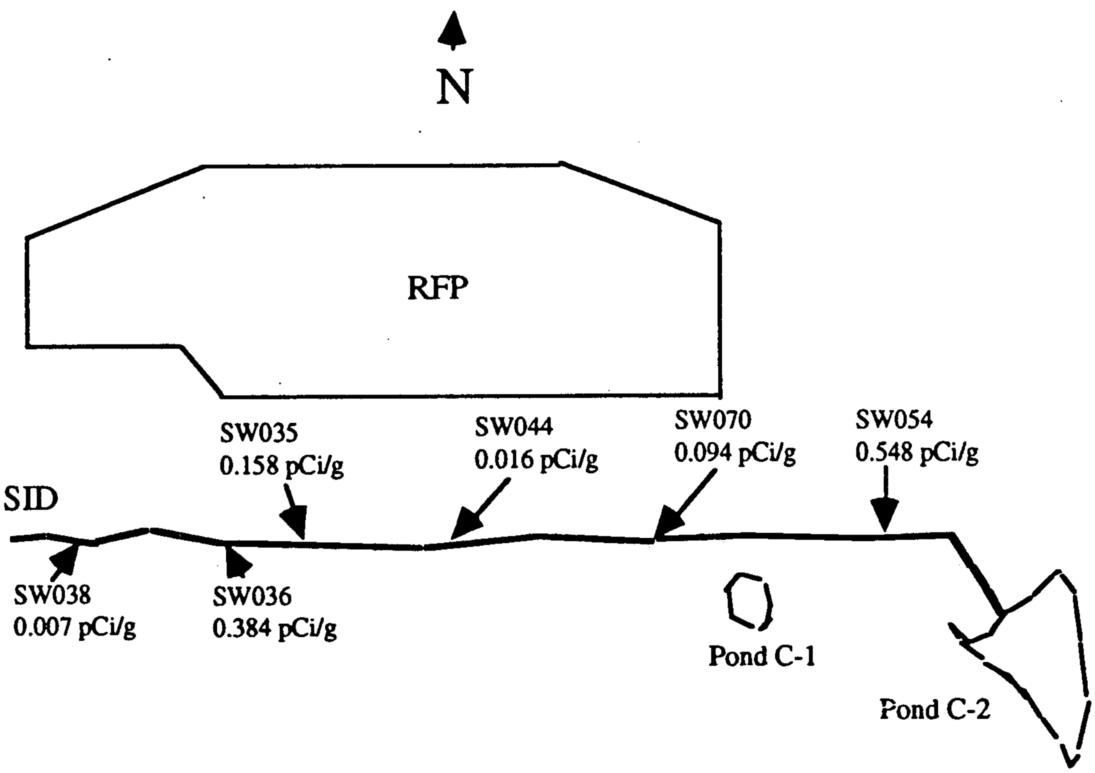
Pond C-2 contained 2.73 pCi $^{239+240}Pu/g$. No plutonium was detected in the sediment collected at the Coal Creek-Woman Creek Headgate. The sediment collected at the Walnut and Indian Sample Pond contained RFP plutonium and anthropogenic uranium. The plutonium content of the sediment sample collected from the Landfill Pond was typical of surface soil samples at RFP.[6] There does not appear to be any evidence that plutonium is accumulating in the Landfill Pond sediments.

Figure 5 is a schematic showing the sampling locations in the South Interceptor Ditch.

Figure 5

Sampling Locations in the South Interceptor Ditch



The plutonium appears to be entering the ditch at specific locations below sampling location SW038. Sample SW60059JE collected at sampling location SW038 contained 0.007 pCi $^{239+240}Pu/g$. The

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plutonium in this sample had a $^{240}Pu/^{239}Pu$ atom ratio of 0.16 $\pm$ 0.03. The plutonium concentration and isotopic composition of SW60059JE are consistent with global fallout.[19,20,21] Plutonium seems to be entering the South Interceptor Ditch in the vicinity of sampling locations SW035 and SW036. Sample SW054 contained the highest plutonium concentration measured in the ditch.

The uranium activities at each sampling location are reported in Table XI and the atom percentages of each uranium isotope are reported in Table XII.

Table XI

Activity of Specific Uranium Isotopes in RFP Sediment and Soil Samples

Sample Number	Description	Total Uranium pCi/g	U-234 pCi/g	U-235 pCi/g	U-236 pCi/g	U-238 pCi/g
SW60038JE	A-1 Bypass Sediments	1.8683	0.9314	0.0372	0.0016	0.8981
SW60277WC	A-1 Pond Sediment	4.3673	1.3284	0.0673	0.0138	2.9578
SW60285WC	A-2 Pond Sediment	6.0060	1.9208	0.1015	0.0178	3.9659
SW60270WC	A-3 Pond Sediment	1.6746	0.7660	0.0355	0.0014	0.8717
SW60268WC	A-4 Pond Sediment	1.1691	0.5387	0.0249	0.0010	0.6294
SW60286WC	B-1 Pond Sediment	8.7425	3.4191	0.1598	0.0320	5.1316
SW60287WC	B-2 Pond Sediment	4.1631	1.9624	0.0788	0.0080	2.1139
SW60276WC	B-3 Pond Sediment	6.5042	2.9439	0.1136	0.0132	3.4335
SW60272WC	B-4 Pond Sediment	1.2606	0.6038	0.0275	0.0011	0.6282
SW60269WC	B-5 Pond Sediment	0.9693	0.4686	0.0211	0.0004	0.4792
SW60026JE	C-1 Pond Sediment	0.5769	0.2824	0.0129	0.0000	0.2816
SW60278WC	C-1 Pond Sediment	1.9502	1.0181	0.0405	0.0003	0.8913
SW60279WC	C-2 Pond Sediment	2.7589	1.1664	0.0508	0.0057	1.5360
SW60035JE	*Coal Creek:Woman Creek	0.7044	0.3626	0.0150	0.0000	0.3268
SW60040JE	Landfill Pond Sediments	0.8500	0.4018	0.0196	0.0000	0.4286
SW60004JE	South Interceptor Ditch	1.1429	0.5028	0.0221	0.0019	0.6161
SW60005JE	South Interceptor Ditch	1.0044	0.4115	0.0198	0.0017	0.5714
SW60006JE	South Interceptor Ditch	0.7308	0.3177	0.0154	0.0007	0.3970
SW60007JE	South Interceptor Ditch	0.8002	0.4040	0.0182	0.0002	0.3778
SW60008JE	South Interceptor Ditch	1.5972	0.5082	0.0279	0.0044	1.0567
SW60009JE	South Interceptor Ditch	0.4754	0.2700	0.0106	0.0006	0.2321
SW60041JE	South Interceptor Ditch	2.5442	1.0602	0.0463	0.0053	1.4324
SW60059JE	South Interceptor Ditch	1.6658	0.8631	0.0345	0.0002	0.7680
SW60288WC	**Walnut and Indian	1.4168	0.6864	0.0295	0.0011	0.6998

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Table XII

Atom Percent of Uranium Isotopes Present in RFP Sediments and Soils

Sample Number	Description	U-234	U-235	U-236	U-238
	Natural Uranium	0.0057	0.7204	0.0000	99.2739
SW60038JE	A-1 Bypass Sediments	0.0056	0.6483	0.0009	99.3452
SW60277WC	A-1 Pond Sediment	0.0025	0.3571	0.0024	99.6380
SW60285WC	A-2 Pond Sediment	0.0026	0.4017	0.0023	99.5933
SW60270WC	A-3 Pond Sediment	0.0048	0.6383	0.0009	99.3561
SW60268WC	A-4 Pond Sediment	0.0047	0.6183	0.0008	99.3763
SW60286WC	B-1 Pond Sediment	0.0036	0.4884	0.0033	99.5047
SW60287WC	B-2 Pond Sediment	0.0051	0.5842	0.0020	99.4087
SW60276WC	B-3 Pond Sediment	0.0048	0.5186	0.0020	99.4747
SW60272WC	B-4 Pond Sediment	0.0052	0.6861	0.0010	99.3077
SW60269WC	B-5 Pond Sediment	0.0053	0.6888	0.0004	99.3054
SW60026JE	C-1 Pond Sediment	0.0055	0.7166	0.0000	99.2779
SW60278WC	C-1 Pond Sediment	0.0062	0.7107	0.0002	99.2829
SW60279WC	C-2 Pond Sediment	0.0041	0.5185	0.0019	99.4755
SW60035JE	*Coal Creek:Woman Creek	0.0060	0.7230	0.0000	99.2709
SW60040JE	Landfill Pond Sediments	0.0051	0.7150	0.0000	99.2799
SW60004JE	South Interceptor Ditch	0.0044	0.5614	0.0016	99.4325
SW60005JE	South Interceptor Ditch	0.0039	0.5426	0.0016	99.4519
SW60006JE	South Interceptor Ditch	0.0044	0.6094	0.0009	99.3853
SW60007JE	South Interceptor Ditch	0.0058	0.7520	0.0003	99.2419
SW60008JE	South Interceptor Ditch	0.0026	0.4145	0.0022	99.5807
SW60009JE	South Interceptor Ditch	0.0063	0.7166	0.0000	99.2771
SW60041JE	South Interceptor Ditch	0.0040	0.5067	0.0019	99.4873
SW60059JE	South Interceptor Ditch	0.0061	0.7026	0.0001	99.2911
SW60288WC	**Walnut and Indian	0.0053	0.6607	0.0008	99.3332

Depleted uranium was detected in the sediments collects from the A and B series ponds. Depleted uranium was also detected in sediments collected from the South Interceptor Ditch, Pond C-2, Walnut and Indian Sampling Pond and the A-1 Bypass. No anthropogenic uranium was detected in sediments collected at the Coal Creek-Woman Creek Headgate, the Landfill Pond or Pond C-2. Anthropogenic uranium appears to be entering the South Interceptor Ditch at specific locations. Table XIII reports the maximum percentage of uranium present in each sediment sample attributable to depleted uranium. The same algorithm was used for water and sediment samples.

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Table XIII

Maximum Percentage of Uranium in Sediments and Soils Attributable to Depleted Uranium

Sample Number	Description	Maximum Percentage of Uranium Attributable to Release of Depleted Uranium From RFP
SW60038JE	A-1 Bypass Sediments	19
SW60277WC	A-1 Pond Sediment	100
SW60285WC	A-2 Pond Sediment	100
SW60270WC	A-3 Pond Sediment	21
SW60268WC	A-4 Pond Sediment	28
SW60286WC	B-1 Pond Sediment	80
SW60287WC	B-2 Pond Sediment	40
SW60276WC	B-3 Pond Sediment	65
SW60272WC	B-4 Pond Sediment	8
SW60269WC	B-5 Pond Sediment	8
SW60026JE	C-1 Pond Sediment	0
SW60278WC	C-1 Pond Sediment	2
SW60279WC	C-2 Pond Sediment	91
SW60035JE	*Coal Creek:Woman Creek	0
SW60040JE	Landfill Pond Sediments	0
SW60004JE	South Interceptor Ditch	48
SW60005JE	South Interceptor Ditch	56
SW60006JE	South Interceptor Ditch	31
SW60007JE	South Interceptor Ditch	0
SW60008JE	South Interceptor Ditch	100
SW60009JE	South Interceptor Ditch	0
SW60041JE	South Interceptor Ditch	68
SW60059JE	South Interceptor Ditch	4
SW60288WC	**Walnut and Indian	15

* Coal Creek: Woman Creek Headgate

** Walnut and Indian Sample Pond

Sediment samples from the ponds and the South Interceptor Ditch were analyzed by gamma spectroscopy. The results are summarized in Table XIV. The ^{40}K concentrations are consistent with previous measurements of naturally occurring radioactive potassium concentrations in surface samples collected in the RFP vicinity and the ^{137}Cs concentrations are consistent with expected radio-cesium concentrations resulting from global fallout.[17,22] Thallium-208, ^{214}Bi, ^{226}Ra and ^{228}Ac are naturally occurring radionuclides that are formed by the radioactive decay of ^{238}U and ^{232}Th. These radionuclides were

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detected in the concentrations expected from the decay of naturally occurring uranium and thorium in the sediment samples. The ^{241}Am concentrations in the sediment samples collected from Ponds B-1, B-2 and B-3 were detectable by gamma spectroscopy. All radionuclides detected in the sediment samples were naturally occurring or the results of global fallout or processes at RFP.

Table XIV

Activities Detected in Sediment and Soil Samples by Gamma Spectroscopy

Sample Number	Location	^{40}K pCi/g	^{137}Cs pCi/g	^{208}Tl pCi/g	^{214}Bi pCi/g	^{226}Ra pCi/g	^{228}Ac pCi/g	^{241}Am pCi/g
SW60277WC	A-1	19.1	0.5	1.9	2.2	2.6	2.0	**ND
SW60285WC	A-2	20.0	1.1	1.9	3.4	3.1	1.7	ND
SW60270WC	A-3	18.6	0.2	3.1	1.1	1.9	2.5	ND
SW60268WC	A-4	21.3	0.1	2.2	2.2	3.8	2.0	ND
SW60286WC	B-1	19.9	0.8	2.5	4.0	2.6	2.1	25.1
SW60287WC	B-2	19.3	0.2	1.8	2.1	3.1	3.0	3.0
SW60276WC	B-3	21.0	0.4	4.1	1.8	2.7	1.2	2.7
SW60272WC	B-4	17.4	0.4	3.7	2.6	2.3	2.2	ND
SW60269WC	B-5	26.0	0.2	2.0	2.7	1.4	1.9	ND
SW60278WC	C-1	15.1	0.9	1.9	2.6	1.9	0.9	ND
SW60279WC	C-2	19.6	0.4	2.7	1.9	2.7	3.1	ND
SW60004JE	*SID	22.9	0.6	3.1	0.3	2.6	2.6	ND
SW60005JE	SID	22.1	0.1	3.0	2.4	2.3	2.5	ND
SW60006JE	SID	17.6	0.7	2.1	1.3	2.9	1.6	ND
SW60007JE	SID	18.1	1.1	2.6	3.3	2.1	1.4	ND
SW60008JE	SID	19.0	0.3	2.2	3.6	1.7	2.1	ND
SW60009JE	SID	24.9	0.6	3.7	2.0	2.4	3.1	ND
SW60041JE	SID	22.1	0.3	3.4	1.7	2.7	1.4	ND

* SID = South Interceptor Ditch

** ND= No ^{241}Am detected in the sample by gamma spectroscopy

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SPECIAL WATER SAMPLES

A series of special water samples was collected. Samples collected at the Raw Water Pond and the B-124 Treatment Facility were collected as part of an effort to locate the source of the depleted uranium detected in the effluent of the Sewage Treatment Plant. The Coal Creek-Woman Creek Headgate water sample was collected to assess water quality upstream from RFP. This sample provides a base-line for the uranium content of water in the creek prior to entry to RFP. The water sample collected at the Land Fill Pond was collected to assess the quality of the water percolating out of the Land Fill. The Boulder Canal sample assesses the quality of water in the canal. The sample collection dates, sample numbers and sampling locations for the special water samples are summarized in Table XV. The uranium and plutonium concentrations measured in the waters are reported in Table XVI.

Table XV

Special Water Sample Collection Sites

Date Sampled	Sample Number	Description
08/12/93	SW60036JE	Raw Water Pond
08/19/93	SW60037JE	OU-4 West Holding Tank
08/12/93	SW60039JE	Landfill Pond
08/12/93	SW60042JE	Coal Creek-Woman Creek Headgate
09/08/93	SW60055JE	Boulder Canal
08/12/93	SW60056JE	Raw Water Pond
09/08/93	SW60057JE	B-124 Treated Water
09/08/93	SW60058JE	B-124 Raw Water

Table XVI

Uranium and Plutonium Concentrations in Special Water Samples

Sample Number	Description	Uranium pCi/g	Plutonium pCi/g
SW60036JE	Raw Water Pond West of Plant	0.38	0.0002
SW60037JE	OU-4 West Holding Tank	78.15	0.0030
SW60039JE	Landfill Pond	2.21	0.0030
SW60042JE	Coal Creek-Woman Creek Headgate	0.77	0.0001
SW60055JE	Boulder Canal	0.42	0.0015
SW60056JE	Raw Water Pond	0.37	0.0002
SW60057JE	B-124 Treated Water	0.10	0.0002
SW60058JE	B-124 Raw Water	0.37	0.0001

No RFP plutonium or anthropogenic uranium was detected in the Raw Water Pond or the influent or effluent waters collected at the potable water treatment facility, B-124. The source of the depleted uranium in the effluent from STP has not been positively identified. No anthropogenic uranium was detected in the water sample collected at the Landfill Pond. The plutonium concentration was at our limit of detection. We found no evidence that the Landfill Pond is affecting the uranium or plutonium contents of surface-waters at RFP. No anthropogenic uranium or RFP plutonium was detected in the water sample collected from the Boulder Canal. We found no evidence that processes at RFP have affected the water in the canal.

The water sample collected from the OU-4 West Holding Tank contained the highest concentration of uranium that we have measured to date. The uranium contained 0.8337 atom percent ^{235}U and 0.0064 atom percent ^{236}U. This indicates that the sample contains enriched uranium. This is the only sample collected to date that contains any enriched uranium.

CONCLUSIONS

The following conclusions are made concerning radioactivity in the water and sediment samples collected at RFP.

The largest source of anthropogenic radioactivity presently affecting surface-waters at RFP is the sediments that are currently residing in the holding ponds. One gram of sediment from a holding pond contains approximately 50 times more plutonium than 1 liter of water from the pond. Two other specific locations have been identified that may affect surface-waters at RFP. Plutonium and depleted uranium appear to be moving down the South Interceptor Ditch and through the A-1 Bypass.

The upper Ponds A-1, A-2, A-3, B-1, B-2, B-3, and B-4 contain measurable quantities of plutonium, americium, and depleted uranium. The plutonium concentrations in these ponds ranged from 0.004 to 3.09 pCi $^{239+240}$Pu/L. The uranium concentrations ranged from 0.2 to 15.8 pCi/L. Essentially 100% of the uranium in Pond A-1 and Pond A-2 originated as depleted uranium. All other ponds, except Pond C-1, contain mixtures of naturally occurring and depleted uranium. No depleted uranium was detected in Pond C-1.

The largest source of radioactivity in the terminal Ponds A-4, B-5 and C-2 was naturally occurring uranium and its decay product radium. There is 70-450 times more alpha activity resulting from the decay of naturally occurring radium than alpha activity resulting from the plutonium in the terminal ponds. Plutonium and americium concentrations in the terminal ponds were consistent with the values published in our previous report. The largest source of anthropogenic radioactivity in the terminal ponds was depleted uranium. Approximately half of the uranium present in Ponds A-4 and C-2 originated as depleted uranium. Approximately 20% of the uranium in the waters collected from Pond B-5 originated as depleted uranium.

All of the radioactivity observed in soil, sediment and water samples collected at RFP was naturally occurring, the result of processes at RFP or the result of global fallout. No extraneous anthropogenic alpha, beta or gamma activities were detected.

Approximately one-third of the uranium present in the effluent from the Sewage Treatment Plant (STP) originated as depleted uranium. No

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depleted uranium was detected in the Raw Water Pond that supplies the water processed at STP. No plutonium or anthropogenic uranium was detected in the influent or effluent from the Water Treatment Facility at B-124. The source of depleted uranium in the effluent from STP has not been positively identified.

No uranium or plutonium attributable to RFP was detected in sediments collected at the Coal Creek-Woman Creek Headgate. Plutonium and depleted uranium were detected in sediments collected from the Walnut and Indian Street Sampling Pond. This indicates that the depleted uranium and plutonium are entering the surface-waters at the RFP site.

Plutonium and depleted uranium were detected in soil samples collected from the South Interceptor Ditch. The plutonium and uranium concentrations varied from location to location within the South Interceptor Ditch. This suggests that these materials are entering the ditch at specific places. The uranium in the sediments collected from Pond C-1 is naturally occurring uranium. Approximately 50% of the uranium detected in the waters and 90% of the uranium detected in the sediment sample collected from Pond C-2 were anthropogenic. This implies that the depleted uranium is being transported down the South Interceptor Ditch by water into Pond C-2.

Radium activities in the water samples were consistent with the activities predicted to be present from the naturally occurring uranium. In general the radium activities were lower than the total uranium activities. This is explained by the fact that the water samples contained depleted uranium. The radium is chemically separated during uranium processing. The depleted uranium released into the environment did not contain large amounts of radium.

The plutonium concentrations in Pond C-2 appear to vary seasonally.

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SUGGESTIONS FOR FURTHER STUDIES

1. This study identified specific locations that affect the radioactivity content of surface-waters at RFP. We suggest that additional locations be characterized in order to determine the affect that they are having on surface-waters. Locations discovered to affect the surface-waters at RFP should be sampled in a manner that provides data that can be modeled to predict further impacts of the site on water quality.

2. To the best of our knowledge this is the first study that has been able to separate the uranium in environmental samples into its naturally occurring and its anthropogenic components. We suggest that sampling efforts be continued to characterize the depleted uranium plume that was discovered at RFP. Samples should be collected on site and off site. Wells and seeps should be monitored to determine if anthropogenic uranium is present. Soil samples should be analyzed to determine if depleted uranium exists as surface contamination.

3. The plutonium concentration in Pond C-2 seem to vary seasonally. Studies should be initiated to determine the cause for this variation. Identification of the cause of the seasonal variation may provide information which is useful in guaranteeing discharges from Pond C-2 always remain in compliance with all appropriate regulations. We recommend that a limnologist be consulted to help plan the appropriate experiments.

4. The plutonium concentration measured in water collected from Pond C-1 on August 23, 1993, was significantly higher than previous measurements. The pond should be monitored to determine if this high plutonium concentration was an anomaly or an indication that plutonium is now being introduced into the pond.

5. The uranium concentration in Pond A-1 seemed to increase throughout this study. Pond A-1 should be monitored to determine if the increase was real or a sampling artifact.

6. The water sample collected from the OU-4 West Holding Tank contained uranium that was enriched in ^{235}U. We suggest that additional samples be collected to verify or repudiate the original measurement.

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Appendix 1

Natural Decay Series

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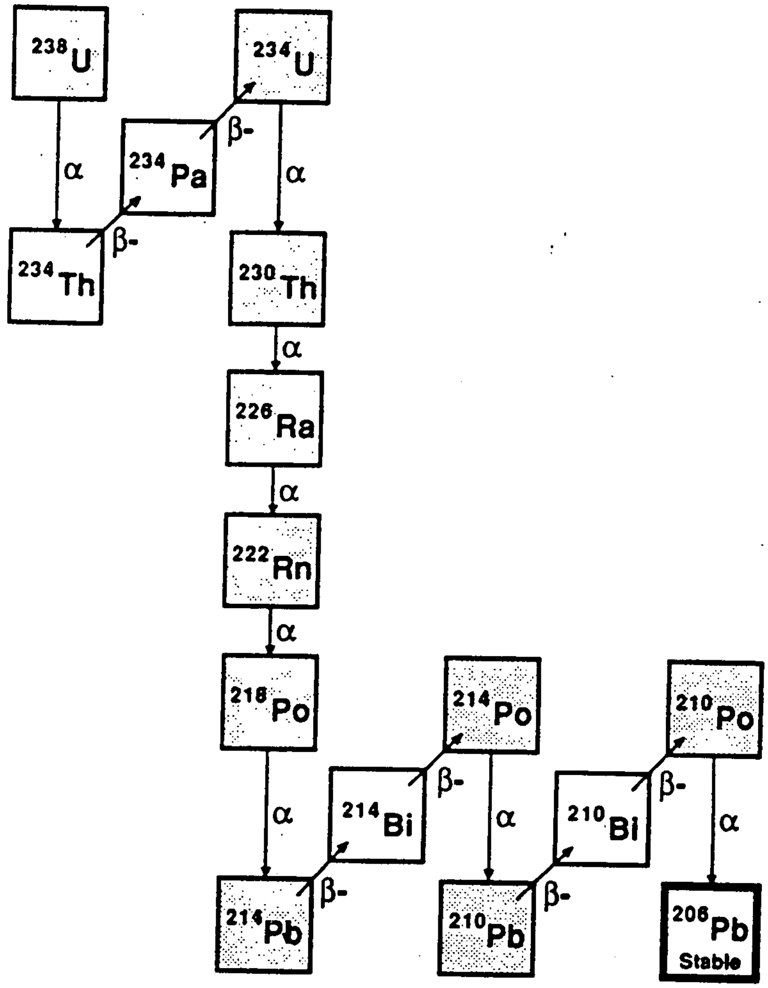
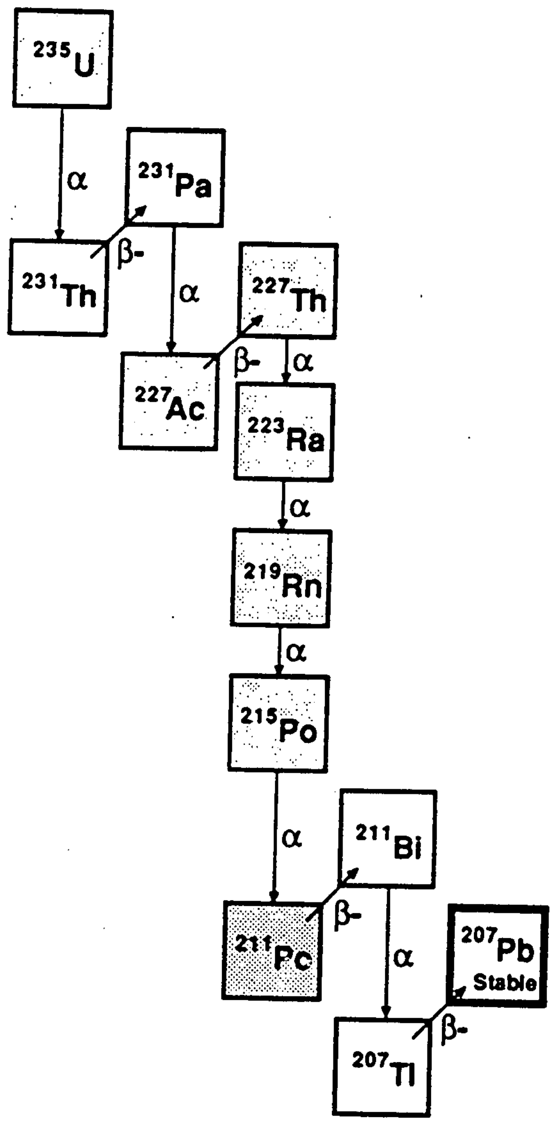


238U
α
234Th
β-
234Pa
β-
234U
α
230Th
α
226Ra
α
222Rn
α
218Po
α
214Pb
β-
214Bi
β-
214Po
α
210Pb
β-
210Bi
β-
210Po
α
206Pb
Stable



^{235}U
α
^{231}Pa
^{231}Th
β-
α
^{227}Th
^{227}Ac
β-
α
^{223}Ra
α
^{219}Rn
α
^{215}Po
α
^{211}Bi
^{211}Pc
β-
α
^{207}Pb
Stable
^{207}Tl
β-

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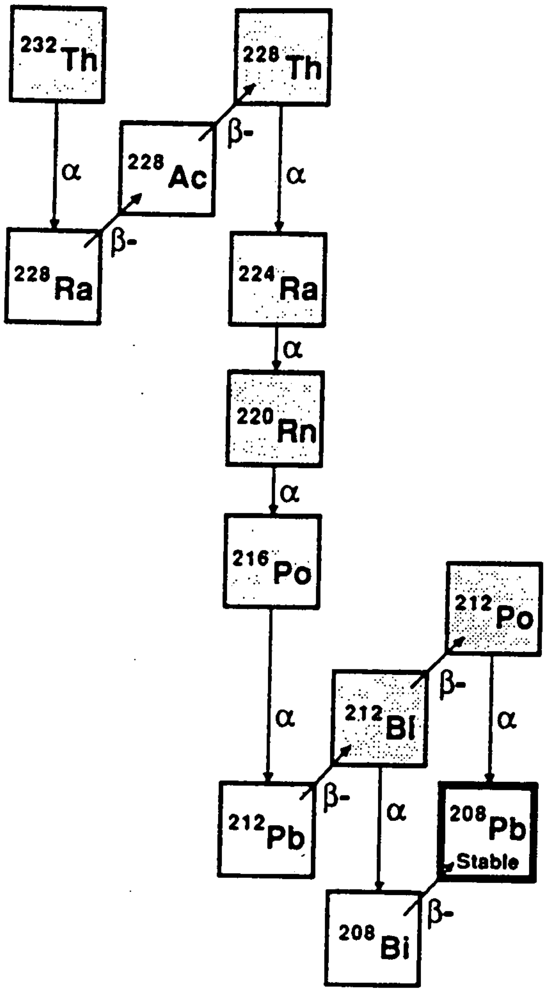
232Th
α
228Ra
β-
228Ac
β-
228Th
α
224Ra
α
220Rn
α
216Po
α
212Pb
β-
212Bi
β-
212Po
α
α
208Bi
β-
208Pb
Stable